Economic and full environmental assessment of electrofuels via electrolysis and co-electrolysis considering externalities

Diego Freire Ordóñez [a,b*], Nilay Shah [b], Gonzalo Guillén-Gosálbez [c]

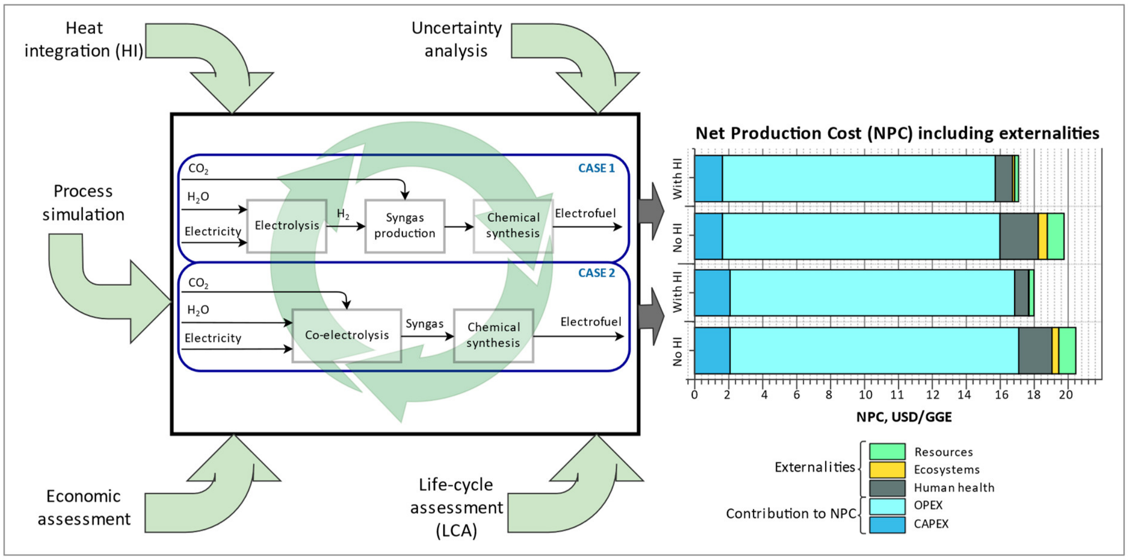
[a] *Institute for Applied Sustainability Research, Av. Granados E13-55 e Isla Marchena, No. 44, Quito 170503, Ecuador*

[b] *Department of Chemical Engineering, Imperial College London, South Kensington, London, SW7 2AZ, UK*

[c] *Institute for Chemical and Bioengineering, Department of Chemistry and Applied Biosciences, ETH Zürich, Vladimir-Prelog-Weg 1, 8093, Zürich, Switzerland*

dmf15@ic.ac.uk

Graphical Abstract



Abstract

Electrofuels from CO_2 and H_2O have recently emerged as a promising alternative to reduce the carbon footprint of fossil fuels, yet their full economic and environmental performance remains unclear. Here, the production of renewable petrol from electrolysis and co-electrolysis-based processes is critically assessed, combining a palette of tools encompassing process simulation, costing evaluation, life-cycle assessment, and uncertainty analysis. Our results show that electrofuels are currently very expensive (10.4-fold higher cost compared to petrol), even when considering externalities (indirect cost of environmental impacts). Electrofuels could become cheaper than the fossil analogue, yet this would require relying on low-cost renewable electricity, which may find alternative uses. From an environmental perspective, we found that despite reducing the carbon footprint of the fossil counterpart, electrofuels could exacerbate impacts on human health due to burden-shifting. Overall, our work highlights the need to embrace impacts beyond climate change to ensure a comprehensive assessment of alternative fuels, and to monetise them to underpin a fair comparison with the fossil analogue.

Keywords: Electrolysis and co-electrolysis, Electrofuels, Techno-economic assessment, Life-cycle assessment, Uncertainty analysis, Monetisation of environmental impacts

1. Introduction

Worldwide energy demand has grown steadily in recent decades, leading to a substantial increase in the consumption of renewable and non-renewable energy sources. To meet this energy demand sustainably, research efforts are focusing on replacing conventional fossil energy with renewables, mainly in four different areas, *i.e.*, power generation, heating, transportation, and rural energy services [1].

At present, transportation is responsible for about 19% of the global energy use and 23% of the energy-related CO_2 emissions, shares projected to increase by approximately 50% by 2030, and by 80% by 2050 [2]. In this context, synthetic fuels based on CO_2 have recently emerged as an attractive alternative to reduce fossil fuels consumption, mitigate climate change and enhance energy security [3,4]

The term “synthetic fuel” applies to a manufactured fuel with approximately the same composition and specific energy as those of a natural fuel [5]. Biofuels converting biomass into high-energy-density fuels initially attracted substantial interest. Competition for food production, deforestation and land-use change [6], however, shifted the focus of recent research towards other renewable sources; these include wind and solar energy, where the excess of electricity due to their intermittency can be harnessed to produce synthetic fuels [7].

Renewable and non-renewable synthetic fuels are being investigated, where the former are classified according to the share of electricity used in their production; specifically, the term “electrofuels” refers to those requiring a large amount of electricity [8]. Electrofuels are carbon-based fuels obtained from CO_2 and water using electricity as the primary energy source to activate the inert CO_2 molecule [9,10]. They could be used to store electricity in chemical molecules, or as feedstock to produce other high-value products [11]. Notably, the so-called renewable electrofuels are considered nearly carbon-neutral concerning greenhouse gas emissions [12], as both the electricity and the carbon are provided by renewable sources [13].

The production of electrofuels has been highly researched during the last years aiming at both mitigating resources deployment and decarbonising the transportation sector [14]. In passenger vehicles, using electricity as the energy source is considered a more sustainable and cleaner option compared to the use of fossil fuels [7]. Unfortunately, in some vehicles, such as ships and long-haul trucks, the required batteries may fail to meet current standards, such as high energy density, a high degree of autonomy, or a brief refuelling time [15]. Hence, electrofuels may find applications in commercial air transport along with long-distance transport, shipping, and the production of carbon-intensive structural materials [12,14], which are hard to electrify. Furthermore, electrofuels can help to curb fossil CO_2 emissions while addressing the issue of intermittency of renewables and its implications on the grid's reliability [16,17].

The deployment of these fuels, however, must still overcome some barriers. In this context, as stated by Albrecht et al. [18], the competitiveness of synthetic fuels on the market will rely on the fuel net production costs, the anticipated cost reduction potential, and the policies for climate change mitigation. Along these lines, Speight [19] claims that synthetic fuels could achieve profitability depending on the feedstock and the production process. Gauging the potential of electrofuels requires performing techno-economic and environmental analyses considering the entire production processes and featuring a similar level of detail and assumptions [18]. In this regard, several studies provide guidelines to conduct techno-economic analyses [18,20], environmental analyses [21–23], or both [24,25], specifically applicable to synthetic fuels. An exhaustive literature review on this topic (see Table 1) reveals that a standardised methodology has not been embraced yet, making it hard to carry out objective comparisons.

Table 1 reviews recent works on liquid electrofuels, most of which have focused on green oxymethylene dimethyl ethers (OME_n), methanol, and Fischer-Tropsch (*FT*) fuels from CO_2 and electrolytic H_2.

Table 1. Literature review of the production of liquid electrofuels through process modelling.

Source	Main liquid products	Electrolysis technology				*FT*	Economic assessment			LCA			Uncertainty analysis		
							Yes		No	Yes		No	Yes		No
		PEM	*SOEC*	*AE*	Not specified	*FT*-based fuels	Without externalities	With externalities		Only *GW* indicators	*GW* + other indicators		Economic uncertainty	Environmental uncertainty	
Samavati et al. [26]	Diesel		x			x	x					x			x
Al-Qahtani et al. [27]	Methanol	x					x				x				x
González-Garay et al. [28]	Methanol	x						x			x		x		
Martín et al. [29]	Methanol				x		x					x	x		
Michailos et al. [24]	Methanol	x					x				x		x		
Zhang et al. [30]	Methanol		x				x					x	x		
Wang et al. [20]	Methanol, gasoline		x				x					x	x		
Albrecht et al. [18]	Liquid fuel	x				x	x					x			x
Alhyari et al. [31]	Liquid fuel		x			x			x		x				x
Albrecht et al. [32]	Liquid fuel		x			x	x					x			x
Becker et al. [33]	Liquid fuel		x			x	x					x			x
Cinti et al. [34]	Liquid fuel		x			x			x			x			x

Source	Main liquid products	Electrolysis technology				*FT*	Economic assessment			LCA			Uncertainty analysis		
							Yes		No	Yes		No	Yes		No
		PEM	*SOEC*	*AE*	Not specified	*FT*-based fuels	Without externalities	With externalities		Only *GW* indicators	*GW* + other indicators		Economic uncertainty	Environmental uncertainty	
Fasihi et al. [35]	Liquid fuel			x		x	x					x			x
Herz et al. [36]	Liquid fuel		x			x	x					x			x
König et al. [37]	Liquid fuel	x				x	x					x	x		
Schemme et al [38]	Liquid fuels	x				x	x					x	x		
Bongartz et al. [39]	OME_1				x				x			x			x
Burre et al. [40]	OME_{3-5}				x				x			x			x
Hank et al. [41]	OME_{3-5}	x					x				x				x
Zhang et al. [42]	Methanol, jet fuel		x			x			x			x	x		

Bearing the above in mind, we focus here on electrofuels obtained via *FT*, which can replace a wide range of fossil fuels without modifying the current infrastructure or motors due to their very similar properties [43]. As seen in Table 1, practically all the existing studies covering environmental assessments of *FT* fuels focused on their global warming potential. However, none of them provides full insight into the environmental impacts associated with their production, *i.e.*, characterisation of impacts at the midpoint and endpoint levels, widely analysed in conventional LCA studies. Hence, the extent to which these fuels can contribute to sustainable development remains unclear, as a full comprehensive LCA encompassing a wide range of impact categories is lacking. Furthermore, these fuels' economic assessments often omit the economic savings linked to lower environmental impacts (relative to their fossil analogues). Notably, reducing impacts results in lower indirect costs (*i.e.*, externalities). However, quantifying these environmental savings is considered critical to ensure a fair comparison, as synthetic fuels from biomass or CO_2 and electricity are currently more expensive than conventional fossil fuels [44]. Indeed, the main motivation for adopting them is environmental rather than purely economic.

In this regard, here we evaluate two technologies to produce electrofuels displaying very similar properties to those of conventional petrol. The technologies assessed combine either a *PEM* or a *SOEC* electrolyser, with an *FT* reactor to produce petrol from CO_2 and renewable energy. As opposed to other works, a full LCA is here conducted for each production process based on the ReCiPe 2016 damage model, which covers 22 midpoint and 3 endpoint indicators. The latter are monetised to uncover the real total cost of each fuel, thereby ensuring a fair comparison against the fossil business-as-usual (*BAU*) alternative.

In the optimistic scenario, *i.e.*, when omitting the impact embodied in wind energy by assuming the use of only surplus energy from intermittent wind, electrofuels outperform fossil petrol in the three endpoint categories of the LCA (human health, ecosystems, and resources). However, they are more expensive to produce than their fossil analogue, even when considering free wind energy. However, the inclusion of externalities would make them economically competitive due to the higher environmental impact of the fossil alternative. However, when the impact of wind is considered, electrofuels lead to burden-shifting toward human health. In the latter case, even when considering externalities, electrofuels are currently not cheaper with existing technology than the fossil alternative.

The article is organised as follows. The assessment methodology, which combines process modelling, heat integration, and techno-economic and life-cycle analyses with the monetisation of impacts, is described first. The results are then assessed and thoroughly discussed, including an extensive comparison against the fossil-based analogue in the UK. Finally, the current work's main findings are highlighted in the conclusions section, along with some recommendations for future work.

2. Methodology

Here we carry out a techno-economic and environmental assessment of two promising electrofuel production processes by combining a palette of tools, as we did elsewhere [28,45–47]. Process simulation is employed to quantify the mass and energy flows associated with fuel production, which are used to evaluate their economic and environmental performance. The former is quantified via the total cost, while to estimate the latter, we apply LCA principles based on the ISO 14040 series (2006). We consider that the plant is located in the UK. A summary of the software, calculation tools, and references from previous studies used in each step of our research is shown in Appendix G, Table G-1.

2.1. Model description

Two flowsheets were built in Aspen Plus v10 [48] based on the conceptual process design presented by König et al. [37] and outlined in Fig. 1 (for the *PEM* electrolysis-based process), and in Fig. 2 (for the *SOEC* co-electrolysis-based process), respectively. This model comprises pure components, *i.e.*, H_2O, CO_2, H_2, CO, and n-alkanes ranging from C_1 to C_{30}. The Peng-Robinson equation of state with Boston-Mathias alpha function (*PR-BM*) was chosen as the reference property method due to its suitability for hydrocarbons [18]. Furthermore, for both electrolysers, the property method electrolyte-NRTL (*ELECNRTL*) was selected to model the electrolysis reactions

[49]. In essence, the electrofuels are produced from syngas generated from CO_2 and H_2 through an *FT* reactor yielding a range of products (depending on the operating conditions and catalysts). We next describe the primary feedstock, then move to the description of the reaction step and, finally, to the separation phase.

2.1.1. Feedstock: Syngas production from CO_2 and H_2

The syngas is a gaseous mixture consisting mainly of hydrogen (H_2), carbon monoxide (CO), and also a certain amount of carbon dioxide (CO_2). This gas, used as intermediate in the production of a wide range of chemicals, including synthetic fuels, currently represents 2% of the total primary energy consumption in the chemical industry [50]. Here, syngas is used as the feedstock of the *FT* process.

In syngas generation, CO is obtained from the reduction of CO_2 through the reverse water-gas shift (*rWGS*) reaction [51]:

$$CO_2 + H_2 \rightleftarrows CO + H_2O \qquad \Delta H = 41.2\ kJ/mol \tag{1}$$

The production of methane often occurs due to two unwanted parallel reactions [52]:

a) CO_2 methanation (known as the Savatier reaction), Eq. (2)

$$CO_2 + 4H_2 \rightleftarrows CH_4 + 2H_2O \qquad \Delta H = -165\ kJ/mol \tag{2}$$

b) CO methanation, Eq. (3)

$$CO + 3H_2 \rightleftarrows CH_4 + H_2O \qquad \Delta H = -206.2\ kJ/mol \tag{3}$$

In fossil-based syngas, methane is used as feedstock, while here we generate the syngas by instead mixing captured CO_2 with electrolytic H_2. Notably, two technologies are studied to produce synthetic syngas, as discussed below, namely electrolysis and co-electrolysis.

PEM electrolysis-based synthesis process (Case 1)

In this technology, electrolytic H_2 is produced first, according to Eq. (4) (see Fig. 1). We modelled the *PEM* electrolyser, including the *PEM* electrolysis unit and its balance of plant (*BOP*), based on the process studied by Michailos et al. [49]. This technology was chosen due to its high *TRL* and market availability [53].

$$H_2O \rightleftarrows H_2 + \frac{1}{2}O_2 \qquad \Delta H = +285.8\ kJ/mol \tag{4}$$

Syngas is obtained next by combining H_2 with CO_2 in an *rWGS* reactor, according to Eq.(1). The *rWGS* reaction requires high temperatures to reach acceptable conversions. Previous works [54–59] studied how to improve the catalytic activity at low temperatures while maintaining a good CO selectivity. Here, a nickel-based catalyst with Al_2O_3 as support [60] is chosen owing to its high activity and competitive cost compared to noble metal-active phase catalysts.

SOEC co-electrolysis-based synthesis process (Case 2)

This technology generates syngas directly from CO_2 and H_2O, according to Eq. (4) and Eq. (5) (see Fig. 2). A *SOEC*, including the electrolysis unit and its *BOP*, was simulated based on the design proposed by Samavati [61]. The motivation for considering the co-electrolysis technology, still under demonstration [53], is two-fold [6]: (i) *SOEC*s may attain larger efficiencies in the future compared to *AE*s and *PEM* electrolysers; and (ii) they can be used for both, electrolysis and co-electrolysis.

$$CO_2 \rightleftarrows CO + \frac{1}{2}O_2 \qquad \Delta H = +283.2 \text{ kJ/mol} \qquad (5)$$

As in the previous case, H_2O and CO_2 undergo an electrochemical conversion to produce syngas, as shown in Eq.(1).

In this work, we followed a simple stoichiometric modelling approach for the electrolysers subsystems, which was previously applied to *PEM*s [49] and *SOEC*s [61]. This approach consists of modelling those subsystems based on the existing reactor types and calculation blocks in Aspen Plus v10, which allow quantifying the mass/energy inputs and outputs, including the electricity demand for the electrolysers under thermoneutral conditions, *i.e.*, the electricity and thermal energy demand of the electrolysis reactions are both covered by electricity [36], here generated from wind power. The electrochemical nature of the electrolysis process is, therefore, not modelled in detail. However, this simplified modelling is consistent with the scope of the analysis and provides a reliable basis for the techno-economic and environmental assessment [49], which is our main focus. We assume an electrical efficiency of 70% for the *PEM* electrolyser [18] and 82% for the *SOEC* [62], and an efficiency of 96% for the AC/DC conversion [37]. To account for the fluctuations of wind power and to guarantee the continuous operation of the synthesis plant, salt caverns are included in the production processes to store syngas [63,64] and hydrogen [37,65] at the exit of the *SOEC* and the *PEM* electrolysers, respectively. Both syngas and hydrogen are produced and stored when wind power is available (considering its full amount, or only the excess above a given value for the more optimistic scenario) and utilised depending on the plant requirements [37,63]. The capacity of the caverns, based on a wind power plant of 46.8% full load fraction, is considered to be 11% of the annual H_2 consumption, and their start-up costs, 5-fold the cost of the cavern [37]. The electricity demand of the chemical plant is covered by grid power in order to ensure its continuous operation.

With regards to CO_2, it can be captured via several technologies [66–68]: (i) Post-combustion, where CO_2 is captured from flue gas streams after combustion; (ii) oxy-fuel, which uses nearly pure oxygen instead of air for fuel combustion; and (iii) pre-combustion, where CO_2 is captured from the reformed synthesis gas of the upstream gasification units.

In this research, post-combustion capture was chosen as capture technology. Data for this process was taken from the work by Irribarren et al. [69], which studied CO_2 capture from coal-fired power plants.

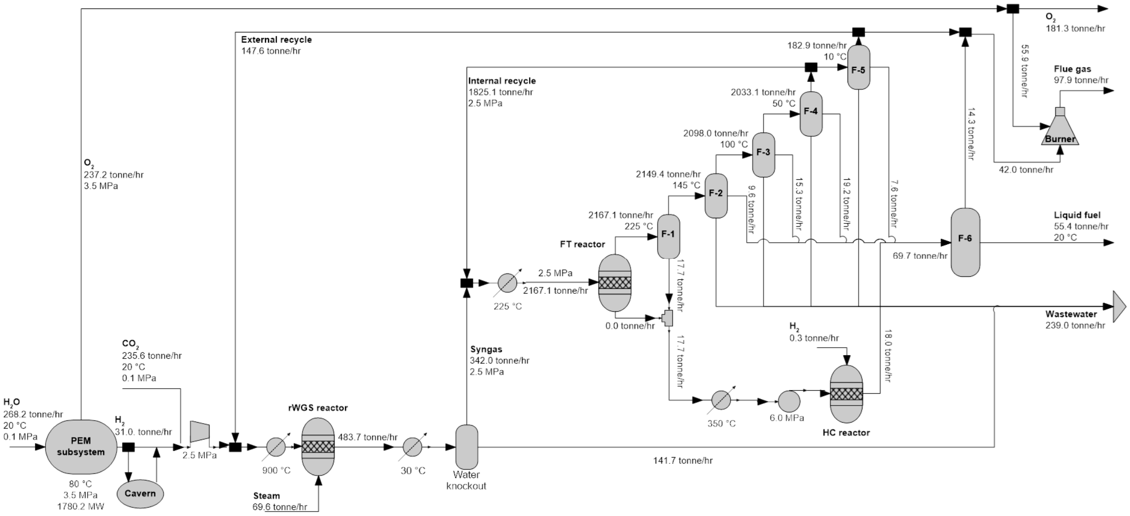


Fig. 1. *PEM*-based process flowsheet (Case 1). The *PEM* subsystem includes the electrolysis unit and the corresponding *BOP* based on [49]

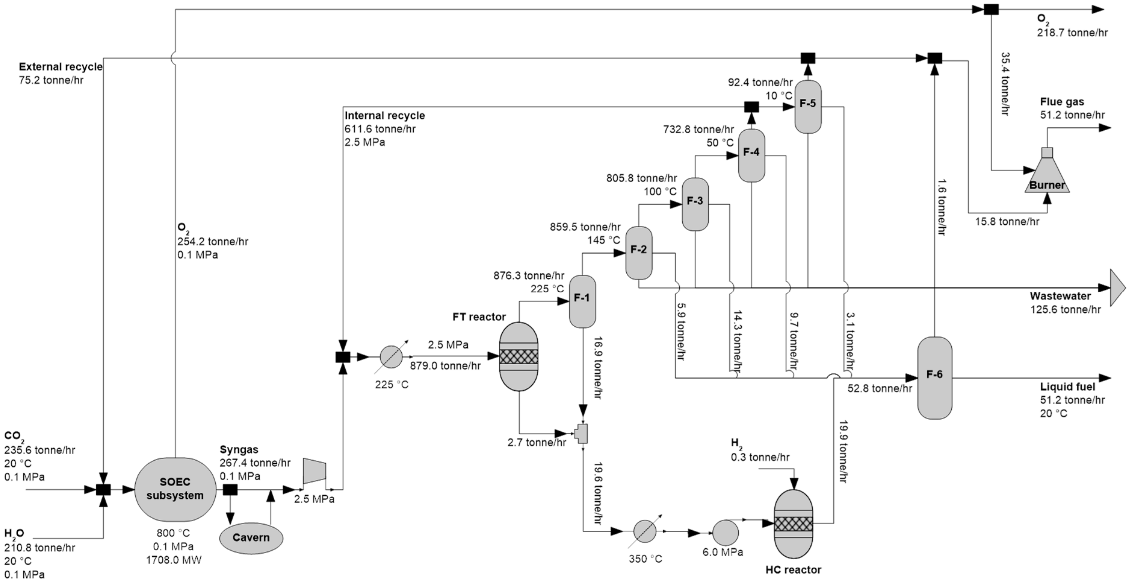


Fig. 2. *SOEC*-based process flowsheet (Case 2). The *SOEC* subsystem includes the electrolysis unit and the corresponding *BOP* based on [61]

2.1.2. Chemical synthesis: Fischer-Tropsch (FT) process

The *FT* process entails a set of polymerisation reactions that transform CO and H_2 into liquid hydrocarbons [70] according to the following reactions:

Hydrogenation of CO to form n-paraffins:

$$nCO + (2n+1)H_2 \rightarrow C_nH_{2n+2} + nH_2O \qquad n=1,2,\ldots,\infty \tag{6}$$

Hydrogenation of CO to form 1-olefins:

$$nCO + 2nH_2 \rightarrow C_nH_{2n} + nH_2O \qquad n=2,3,\dots,\infty \tag{7}$$

Hydrogenation of CO to form oxygenates:

$$nCO + 2nH_2 \rightarrow C_nH_{2n+2}O + (n-1)H_2O \qquad n=1,2,\dots,\infty \tag{8}$$

The ratio of two successive reaction rates is here assumed to follow a constant growth factor known as the chain growth probability, α. This factor is connected via the Anderson–Schulz–Flory distribution to the weight fraction, w_n, which determines the distribution of the products, as follows: [70]

$$w_n = n(1-\alpha)^2\alpha^{n-1} \tag{9}$$

Where n denotes the carbon number or chain length.

In terms of catalyst, iron and cobalt are the preferred choices for *FT* industrial applications, while nickel and ruthenium could also be used, mainly when the focus lies on the production of high molecular weight hydrocarbons [61]. Accordingly, a cobalt-based catalyst [71] is considered herein. The specific *FT* design parameters used in this study [37] are given in Appendix B, Fig. B-1.

2.1.3. Wax hydrocracking (HC)

HC is a catalytic cracking process that is often combined with the *FT* process. Here, long hydrocarbon chains are broken and rearranged while adding H_2 to aromatics and olefins to produce naphthenes and alkanes [72]. This process is regarded as a primary petroleum refining method for the conversion of heavy hydrocarbons into gasoline and middle distillate. Indeed, the middle distillate currently available on the market is mostly produced by crude oil hydrocracking [73]. The products of *HC* are saturated hydrocarbons, from ethane and LPG to heavier hydrocarbons (mostly isoparaffins), depending on the reaction temperature, pressure, and catalyst activity [72].

The hydrocracking of *FT* wax, a long paraffinic chain ($>C_{21}$) [74] obtained in the *FT* process, has been extensively investigated for middle distillate production. *FT* wax possesses several advantages over conventional crude oil as a feedstock for hydrocracking, such as the possibility of producing first-rate middle distillate with low content of sulphur and aromatic compounds [73]. Metal/acid bifunctional catalysts are commonly used, among which NiMo and NiW catalysts supported on solid acids are particularly suitable for middle distillate production [73]. Nevertheless, considering that the *FT* wax is a sulfur-free product, Pd and Pt-loaded catalysts could also be used, both showing excellent performance owing to their elevated hydrogenation/dehydrogenation activity for heavy hydrocarbon cracking [74–76]. Hence, a platinum-based catalyst [77] is considered in this study. The yield distribution considered in the hydrocracker model [37] is given in Appendix B, Fig. B-1. For the required H_2, we consider H_2 coming from wind power produced elsewhere.

2.1.4. Product separation and upgrading

The resulting *FT* gas and the product from hydrocracking should undergo a distillation/flash separation to obtain fuels with similar properties to those of conventional fuels. Hence, six flash separators are considered in the separation step, as shown in Fig. 1 and Fig. 2.

2.2. Heat integration (HI)

Process integration based on mass/recycle targeting, minimum heating, and cooling utilities, and cogeneration targets can be applied to improve further the environmental performance of the process [78]. In our case, the potential energy-savings due to *HI* are estimated using the pinch technology as implemented in Aspen Energy Analyzer v10 [79]. Once the flowsheets are modelled, and utilities are assigned, the software constructs the composite curves and calculates the energy targets. A heat recovery temperature approach of 10°C was selected in both technologies. Each production process is assessed with and without *HI*, and the potential savings are then factored in the economic assessment.

2.3. Economic analysis

2.3.1. Net production cost (NPC)

The NPC of the electrofuels is obtained from the capital (*CAPEX*) and operational (*OPEX*) expenditures, which are calculated using the Aspen Process Economic Analyzer v10 [80]. The installed costs of the electrolysers and reactors were estimated following the guidelines given by Albrecht et al. [18]. The purchased cost is obtained from Eq. (10), using the data in Appendix A, Table A-2. All costs, including raw materials, utilities, products, and equipment, were updated to 2018. In contrast, the cost of the electrolysers, *i.e.*, total direct costs accounting for purchased costs, installation, piping, civil works, steel, instrumentation, electricals, insulation, and paint (see Table A-2), were estimated based on their projected cost to 2020 [62] and the most recent annual plant cost index available at the moment, *i.e.*, $CEPCI_{2019}$ = 607.5 [81]. The chemical engineering plant cost index (*CEPCI*), which was first introduced in 1963, is a regularly-updated composite index obtained from different sub-indexes related to equipment, construction, labour, buildings and engineering costs. It is commonly utilised to adjust the purchased costs of equipment from one period to another [81].

$$\mathrm{PC} = \mathrm{PC}_{\mathrm{ref}} \cdot \left(\frac{\mathrm{S}}{\mathrm{S}_{\mathrm{ref}}}\right)^{\mathrm{D}} \cdot \left(\frac{\mathrm{CEPCI}_{2018}}{\mathrm{CEPCI}_{\mathrm{ref}}}\right) \tag{10}$$

Here the notation used is as follows: *PC* denotes the purchased cost of the equipment, PC_{ref} represents the same purchased cost expressed in monetary units of the reference year, *S* denotes the scale for the capacity in 2018, S_{ref} expresses the reference scale, and *D* is a scaling factor. The chemical engineering plant cost index for 2018 ($CEPCI_{2018}$) is assumed to be equal to 603.1, for 2018 [82].

With the purchased costs, Aspen Process Economic Analyzer estimates the fixed capital investment (*FCI*), as well as the total operation cost (*TOC*), based on the data presented in Appendix A. The *FCI* includes the cost of the purchased equipment and its installation, as well as other related costs, such as piping, civil works, steel, instrumentation, electricals, insulation, paint, general and administrative overheads, contract fees, design, engineering and procurement, and contingencies (see Fig. 4). The *TOC* considers the cost of raw materials, utilities, revenues for byproducts, and other operating costs, such as operating labour cost, maintenance cost, operating charges, plant overhead, and general and administrative expenses (see Fig. 5).

The annualised capital cost (*ACC*) is then calculated from Eq. (11), with the data in Appendix A, Table A-1.

$$\text{ACC} = \text{FCI}\cdot\frac{i\cdot(1+i)^t}{(1+i)^t-1} \tag{11}$$

Where *i* denotes the interest rate and *t*, the plant lifetime.

Finally, the total annualised cost (*TAC*) is calculated by summing up the *TOC* and the *ACC*, while the net production cost (*NPC*) of the electrofuels is calculated by dividing the *TAC* by their annual production (*AP*), as shown in the equations below:

$$\text{TAC} = \text{TOC} + \text{ACC} \tag{12}$$

$$\text{NPC} = \frac{\text{TAC}}{\text{AP}} \tag{13}$$

The *NPC* of electrofuels is expressed in USD per gasoline gallon equivalent (*GGE*), a standard indicator widely used to compare alternative fuels against conventional gasoline. Hence, this metric quantifies the mass of fuel required to equal the energy output of one gallon of liquid gasoline [37]. Therefore, this indicator enables a more consistent comparison between the *NPC* of the produced electrofuels and conventional petrol (see Table B-1).

2.3.2. Uncertainties in cost data

A sensitivity analysis was carried out to identify the most critical variables in the economic assessment. Monte Carlo sampling was applied next to study the impact of changes in these variables on the outcome of the analysis, modelling the main contributors towards the *CAPEX* and *OPEX* expenditures via a normal distribution with a standard deviation of 20% [83].

2.3.3. Abatement cost of carbon emissions

The abatement cost of carbon emissions, that is, the carbon tax that would make electrofuels match the production cost of their fossil-based analogue, can be determined from the fuels' costs and carbon footprint as follows. First, the global warming potential (expressed in $\text{kg}_{\text{CO}_2\text{-eq}}\ \text{kg}_{\text{fuel}}^{-1}$) of the fuel is quantified from the LCA at the midpoint level. Then, the carbon tax that should be established so that the electrofuel and conventional petrol would both display the same total cost is calculated by Eq. (14)

$$\text{Carbon tax} = \left|\frac{\text{NPC}_{\text{electrofuel}}-\text{NPC}_{\text{petrol}}}{\text{GWP}_{\text{electrofuel}}-\text{GWP}_{\text{petrol}}}\right| \tag{14}$$

Where *GWP* and *NPC* denote the global warming potential and net production cost, respectively.

2.4. Life-cycle assessment (LCA)

The standard LCA methodology [84] is applied to quantify the environmental impact of the different fuels, as described in detail next. All the calculations were implemented in SimaPro v9.0 [85] interfacing with Ecoinvent 3.5.

2.4.1. Goal and scope definition

A cradle-to-gate scope is considered, under the assumption that both fuels will be eventually combusted; hence, including other downstream processes would add no discriminatory power to the analysis. The functional unit is 1 kg of electrofuel. The choice of this functional unit was made following standard guidelines defined for carbon capture and utilisation (CCU) projects, which recommend mass-based functional units due to their widespread use for trading chemicals, materials and minerals [86]. Likewise, as shown in Appendix B, the composition and properties of the produced e-petrol and fossil counterpart are assumed to be very similar, which further justifies the choice of our functional unit as the basis for comparison. Ideally, the same analysis could be done by defining as the functional unit the energy content (*i.e.*, 1 MJ) or, even better, a given distance travelled (*i.e.*, one km travelled, which would consider the engine efficiency of each fuel type). However, these alternative functional units would require additional process- and vehicle-dependent data.

As shown in Fig. 1 and Fig. 2, a fraction of the external recycle, which contains gaseous hydrocarbons and unreacted species, is combusted in the burner to generate energy. Likewise, a portion of the produced oxygen is utilised as an oxidising agent (for 20%-excess oxygen combustion). Hence, we consider natural gas and compressed air to be avoided products, as shown in Appendix D, Table D-1. The environmental burdens are economically allocated among the produced oxygen and the electrofuels, depending on the assumed cost of the wind power, *e.g.*, at zero-cost electricity, 26%-74% and 29%-71%, respectively, for the *PEM* and the *SOEC* cases, while at 0.16 USD/kWh, the resulting allocation percentages are 5%-95% and 4%-96%, respectively. The impact embodied in the caverns, the *PEM* and *SOEC* electrolysers was omitted, as it was shown that their contribution to the total impact is rather low [87–89].

2.4.2. Life-cycle inventory (LCI)

The *LCI*s of elementary flows are obtained by combining the mass and energy flows of the foreground system with data of the background system. The latter includes those surrounding processes providing inputs (raw materials and utilities) to the main process. Information on the foreground system is retrieved from the simulation model of the primary process implemented in Aspen Plus. In contrast, background data are retrieved from Ecoinvent (see Appendix D, Table D-1). The *LCI* of the conventional low-sulphur petrol (*BAU*) is directly taken from Ecoinvent 3.5. The allocation at the point of substitution (*APOS*) system model is considered for all inputs and outputs of the production processes.

2.4.3. Environmental impact assessment (EIA)

The ReCiPe 2016 [90] method is applied as implemented in SimaPro, following a hierarchist perspective [91] and focusing on the following midpoints indicators: global warming on human health, stratospheric ozone depletion, ionising radiation, ozone formation on human health, fine particulate matter formation, human carcinogenic toxicity, human non-carcinogenic toxicity, water consumption on human health, mineral resource scarcity, fossil resource scarcity, global warming on terrestrial ecosystems, global warming on freshwater ecosystems, ozone formation on terrestrial ecosystems, terrestrial acidification, freshwater eutrophication, marine eutrophication, terrestrial ecotoxicity, freshwater ecotoxicity, marine ecotoxicity, land use, water consumption on terrestrial ecosystems, and water consumption on aquatic ecosystems. These are aggregated into three endpoints, *i.e.*, human health, ecosystems, and resources.

2.4.4. Uncertainty analysis of environmental data

Following standard practice, uncertainties in the *LCI* entries are evaluated through Monte Carlo simulations in SimaPro. The pedigree matrix is applied to determine the parameters of the underlying lognormal distributions that model the *LCI* elementary flows (*i.e.*, feedstock, emissions, and waste) [92].

2.5. Monetisation of endpoints (externalities)

Endpoint impact categories are monetised according to the methodology proposed by Weidema [93], as already done in previous works [45,94]. In essence, this approach translates the LCA-endpoint impact categories into monetary units through the application of economic penalties to each indicator [93]:

- The economic penalty factor for human health, *i.e.*, 7.4×10^4 EUR_{2003} per 1 DALY, is equivalent to the undiscounted willingness-to-pay value for a life year under small risk increase from involuntary exposure.
- For the ecosystems quality indicator, the conversion factor, *i.e.*, 9.5×10^6 EUR_{2003} per 1 lost species.year, was derived from a single Japanese choice model, and it is equivalent to a 2%-expenditure of the potential income. For the uncertainty, the low and high-range values are given by the Externe study and an experiment in which the 10% protection target from the Convention of Biological Diversity is employed, respectively.
- The monetary factor for the resources depletion indicator, *i.e.*, 8.62×10^{-1} EUR_{2003} per 1 USD_{2000}, was obtained from a forecast of future energy prices, based on the marginal costs of oil production technologies at the time of the study and assuming that the predicted long-term energy costs would not exceed 0.023 EUR_{2003}/MJ (80 USD_{2000}/barrel).

3. Results and discussion

We start by analysing the economic performance of the fuels to then focus on their environmental assessment.

3.1. Process model results

A summary of the primary energy and mass flows of the production processes is given in Table 2. The product composition profiles (Fig. B-1), along with some estimated properties (Table B-1), can be seen in Appendix B.

Table 2. Summary of energy and mass flows from the simulation of the production processes.

Energy flows				**Mass flows**			
Streams	**Case 1: *PEM***	**Case 2: *SOEC***	**Unit**	**Streams**	**Case 1: *PEM***	**Case 2: *SOEC***	**Unit**
	Value				**Value**		
Inputs:				**Inputs**			
Electricity:				CO_2	235.59	235.59	t/h
Electrolyser	1780.20	1708.02	MW	H_2O	337.75	210.80	t/h
Auxiliaries	75.33	75.46	MW	H_2	0.27	0.30	t/h
Other utilities:							
Cooling water	243.50	182.30	MW				
Propane refrigeration	9.69	7.23	MW				

Outputs:				**Outputs:**			
Electrofuel	679.47	619.29	MW	Electrofuel	55.43	51.16	t/h
				Oxygen	181.32	218.73	t/h
				Flue gas	97.88	51.19	t/h
				Wastewater	238.98	125.61	t/h

3.2. Heat integration

Fig. 3 shows the composite curves of both processes. In both cases, the heating demand can be fully satisfied with internal heat exchange. For the non-heat integrated processes, this demand would amount 397.8 MW for the *PEM*-based case, and 357.6 MW for the *SOEC*-based case. Cooling requirements can be reduced to 253.2 MW and 189.5 MW, respectively, for the *PEM* and *SOEC* cases, compared to 650.9 MW and 547.1 MW in the base case without heat integration. This analysis, therefore, shows that the heating and cooling savings for the *PEM* and *SOEC* cases are quite significant (*i.e.*, 39.4 MUSD and 5.2 MUSD, and 31.5 MUSD and 5.1 MSUD, respectively).

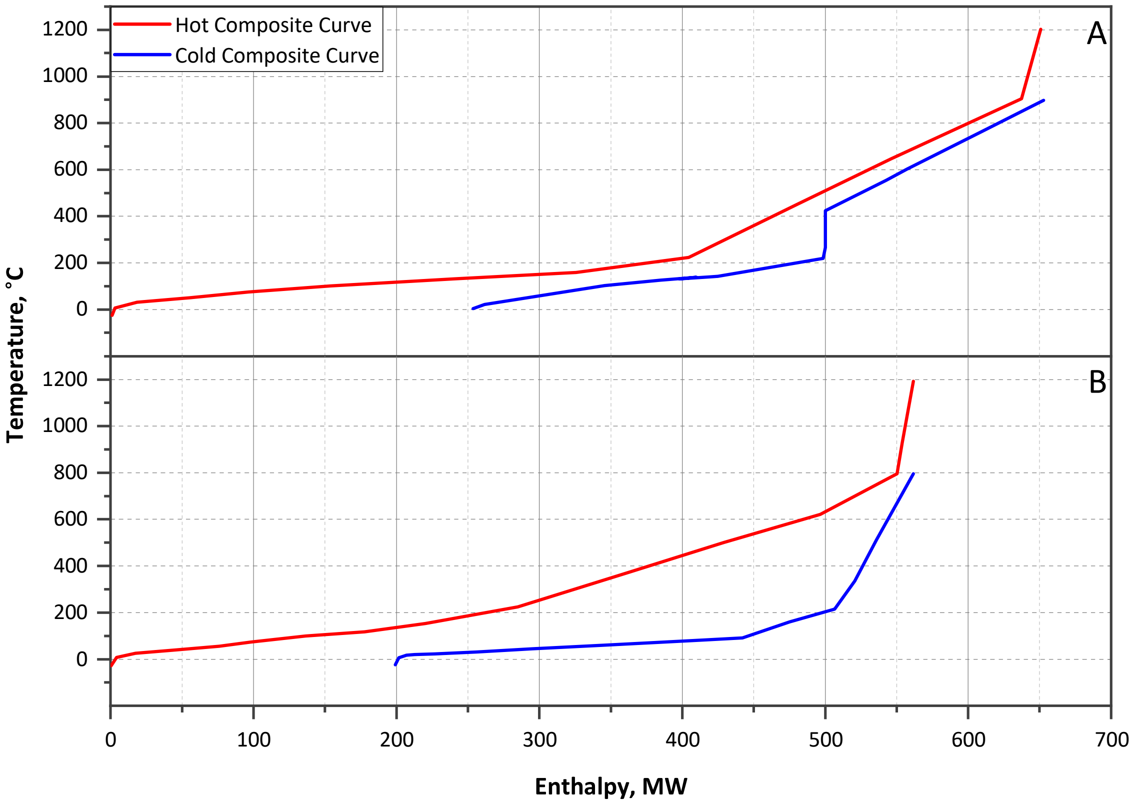


Fig. 3. Composite curves: A. *PEM*-based process; B. *SOEC*-based process.

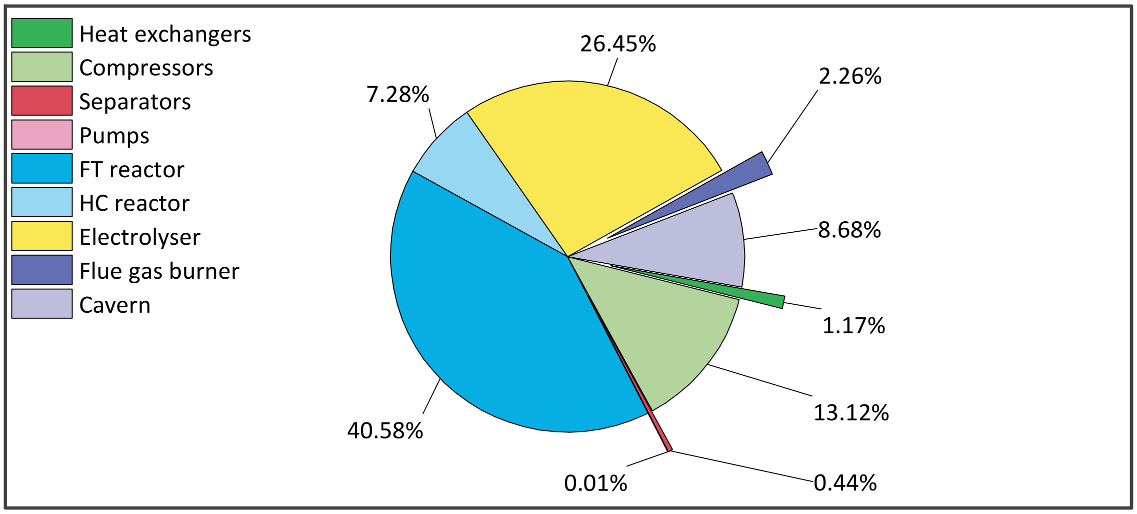
3.3. Economic analysis

Two different scenarios are considered in the economic assessment. The first assumes free wind electricity, as a potential utilisation of the surplus electricity linked to the fluctuating wind power, which needs to be re-dispatched to avoid the stressing of electric grids [95,96]. The second considers the cost of wind power to be 0.16 USD/kWh, which is currently the average cost of offshore wind power in the UK [97].

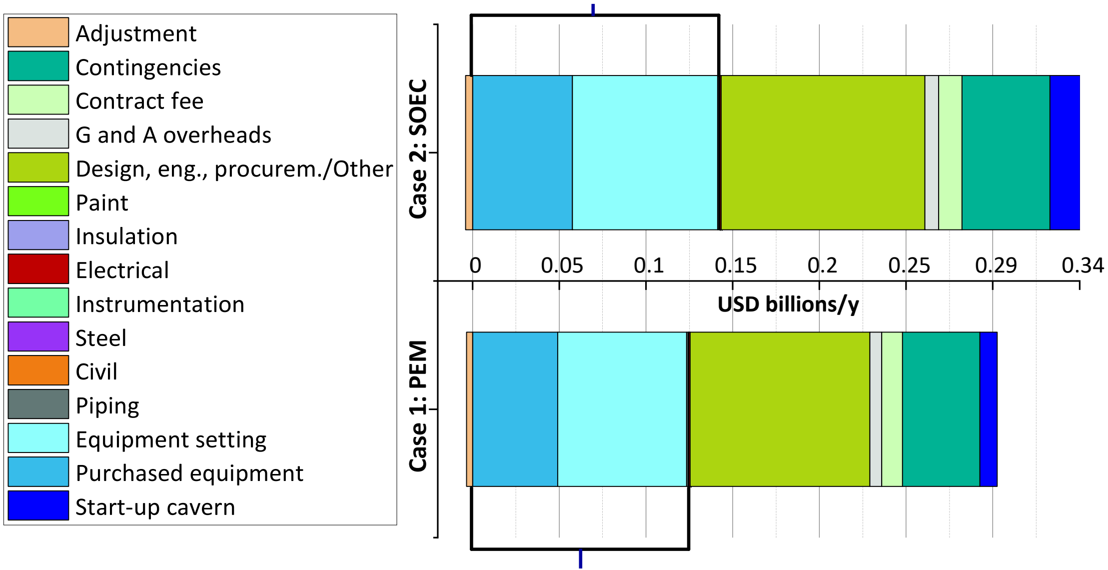
3.3.1. Annualised costs

The annualised costs are summarised in Fig. C-1, whereas the corresponding breakdowns are given in Fig. 4 and Fig. 5. The *PEM* and the *SOEC*-based cases show similar *ACC*. The *TOC* calculated for the actual price of wind power is approximately 15-18-fold higher compared to that estimated under the assumption of zero-cost electricity (see Fig. 5). As expected, the application of heat integration reduces the *TOC*, especially when zero-cost wind power is considered, *i.e.*, a nearly 22%-reduction when heat integration is applied. However, when the actual price of electricity is considered, the potential reductions are in the range of 1.4 to 1.7%. For free wind electricity, the *TOC* of the *PEM*-based process is approximately 23% higher than that of the *SOEC*-based process, compared to only 5% above for an electricity price of 0.16 USD/kWh [97].

With regards to the *CAPEX* (see Fig. 4), its main contributor in terms of equipment is always the *FT* reactor, followed by the electrolyser. Meanwhile, the most significant contributor to the *OPEX* is the captured CO_2, for the case of free electricity, and wind electricity, when its cost is accounted for (see Fig. 5).



Case 2: SOEC. Installed equipment cost distribution



Case 1: PEM. Installed equipment cost distribution

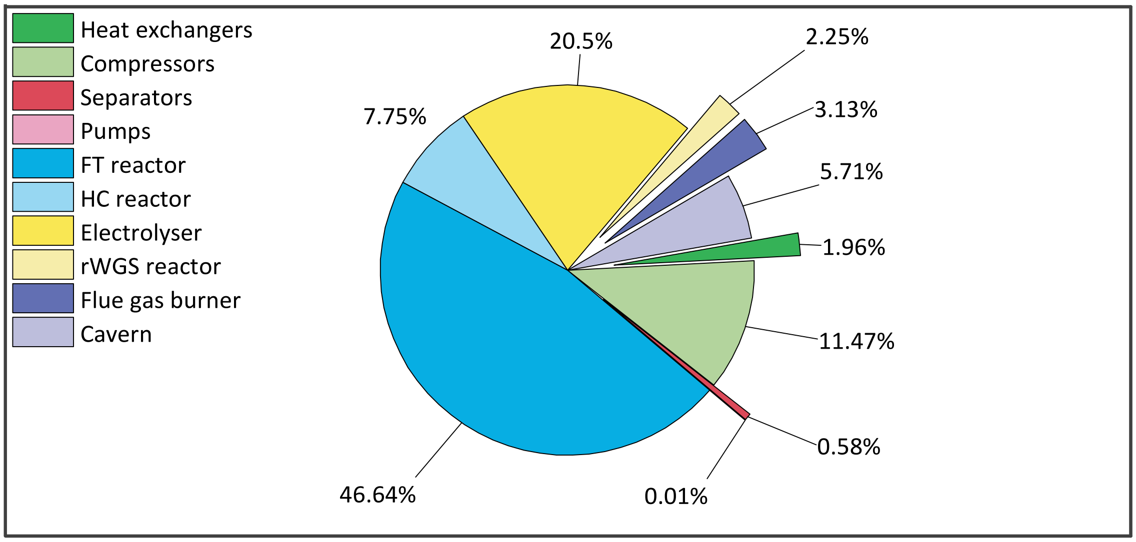


Fig. 4. Breakdown of the annualised capital costs (*ACC*) associated with the electrofuels.

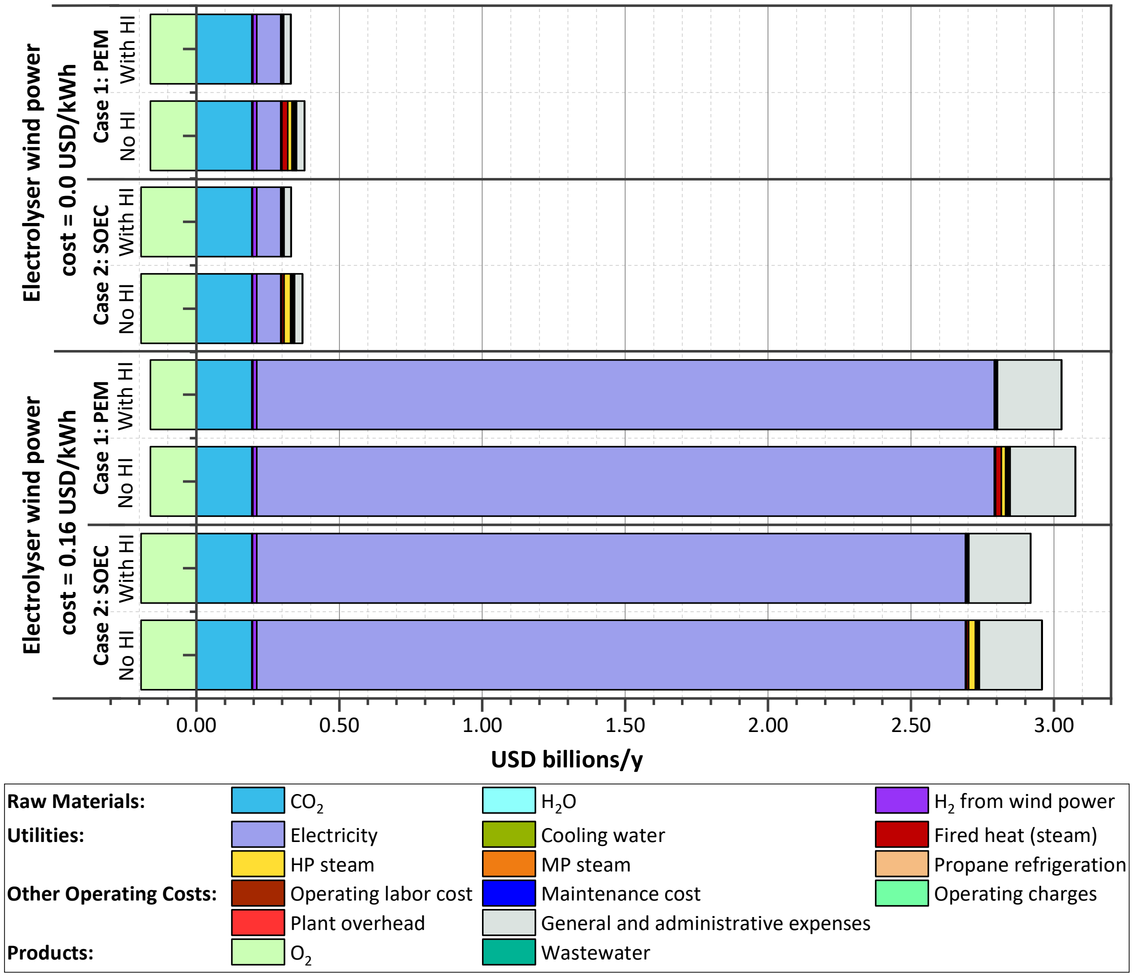


Fig. 5. Breakdown of the total operation cost (*TOC*).

3.3.2. Net production costs (NPC)

The *NPC*s are given in Fig. 6. The externalities shown in Fig. 6A include the environmental impacts of the wind power required by the electrolysers, while in Fig. 6B, those impacts are omitted. The latter case assumes that only surplus electricity from wind is utilised and its corresponding impacts are allocated to the electricity supplied to the grid line (see 3.4). For the heat integrated processes, excluding externalities and assuming zero-cost wind electricity, conventional petrol emerges as the cheapest option (approximately 1.71 USD/gal [98]), followed by the *PEM*-based fuel (2.60 USD/GGE), and then by the *SOEC*-based fuel (2.94 USD/GGE). As shown in Fig. 6A, when externalities are considered, however, the electrofuels outperform the fossil petrol, with the *PEM*-based fuel showing the lowest cost (3.67 USD/GGE vs 3.81 USD/GGE, for the *PEM* and the *SOEC*-based fuels, respectively, and 4.04 USD/gal for the petrol). Accounting for the cost of wind electricity increases the cost of the electrofuels sharply, from 2.60 to 17.73 USD/GGE for the *PEM*-based fuel, and from 2.94 to 18.87 USD/GGE for the *SOEC*-based fuels, making them much more expensive than fossil petrol, even when considering externalities (19.10 USD/GGE vs 20.00 USD/GGE, for the *PEM* and the *SOEC*-based fuels, respectively). The same conclusions stand for the case when the environmental impacts of wind power are omitted for the calculation of the externalities (Fig. 6B).

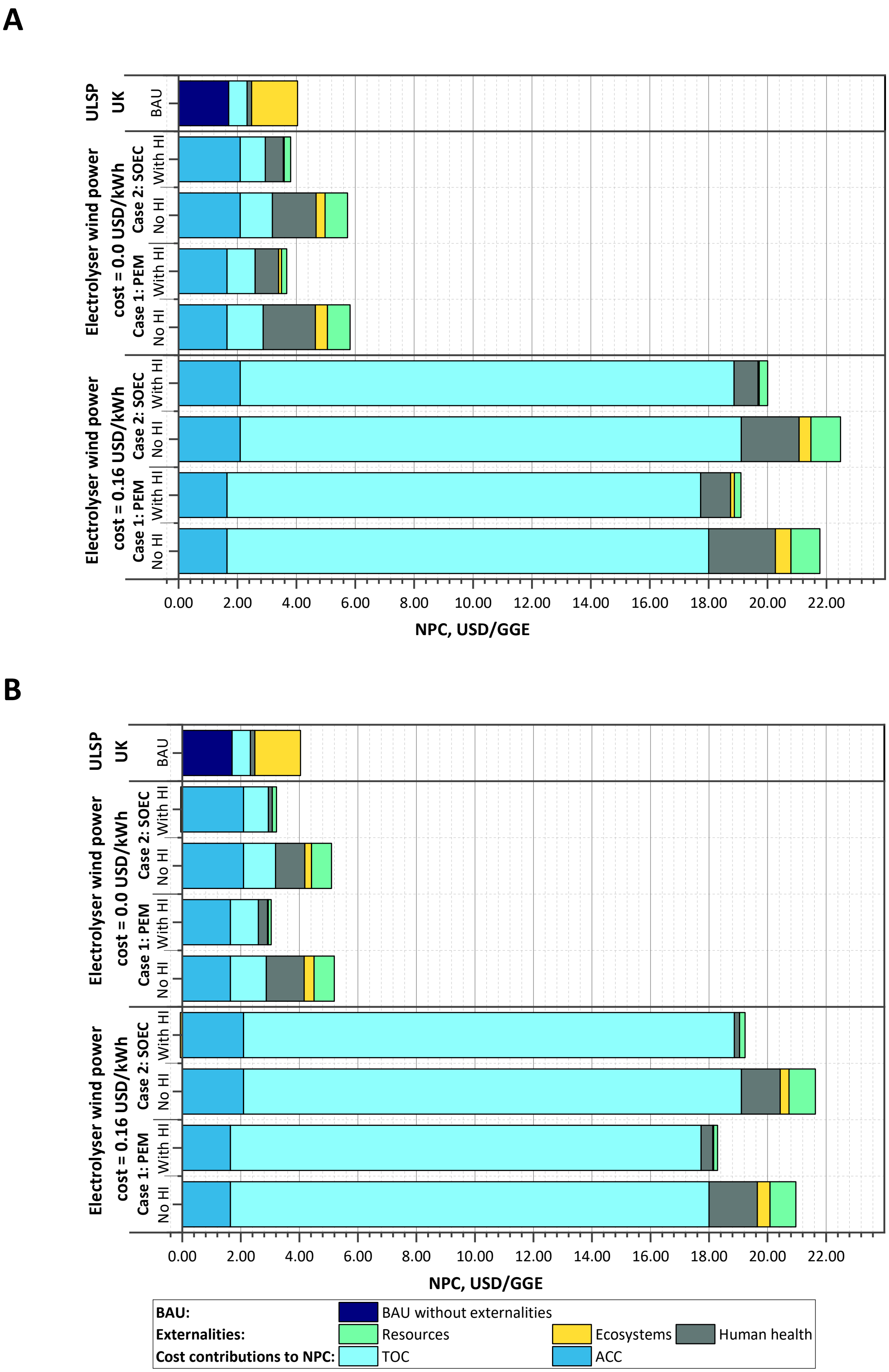


Fig. 6. *NPC* including externalities. A: Externalities including the environmental impacts embodied in wind power; B: Externalities omitting the environmental impacts embodied in wind power. *GGE* stands for gasoline gallon equivalent

3.3.3. *Uncertainty analysis*

The sensitivity analysis, conducted for the NPCs of the electrofuels without externalities, reveals that the cost of wind electricity and, to a lesser extent, the CO_2 cost and the potential selling price of oxygen are the most critical parameters in the economic assessment. The results of the Monte Carlo assessment are shown in Appendix C, Table C-1, and Fig. C-2. The standard deviation of the NPC is 2.95 USD/GGE for the *PEM*-based fuel, and 3.27 USD/GGE for the *SOEC*-based fuel, while the coefficients of variation are 16.67% and 17.42%, respectively.

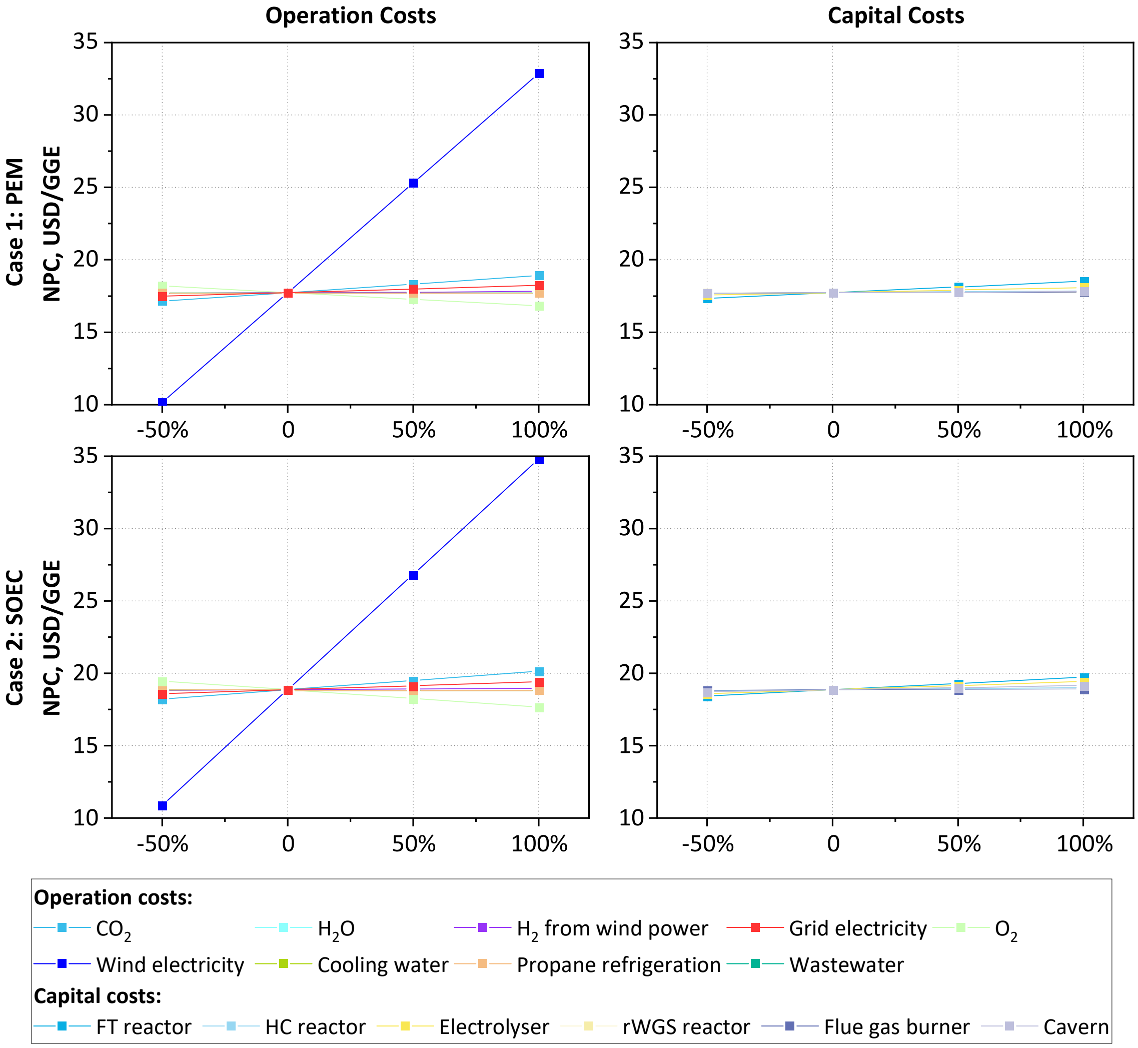


Fig. 7. Sensitivity analysis of cost parameters; electrolyser wind power cost: 0.16 USD/kWh.

3.3.4. *Abatement cost of carbon emissions*

The abatement costs vary in the range 169.98-2587.85 USD $tCO_2\text{-eq}^{-1}$, when the wind power environmental impacts are omitted, and 198.60-3286.45 USD $tCO_2\text{-eq}^{-1}$, when those impacts are assessed, depending on the case and excluding externalities (see Appendix F). We note that these values lie above the current estimated social cost of carbon emissions, *i.e.*, 62.35 USD $tCO_2\text{-eq}^{-1}$ [99]. The abatements costs, *i.e.*, the estimated carbon taxes, along with the *NPC*s of the fuels, are shown in Fig. 8 for each scenario.

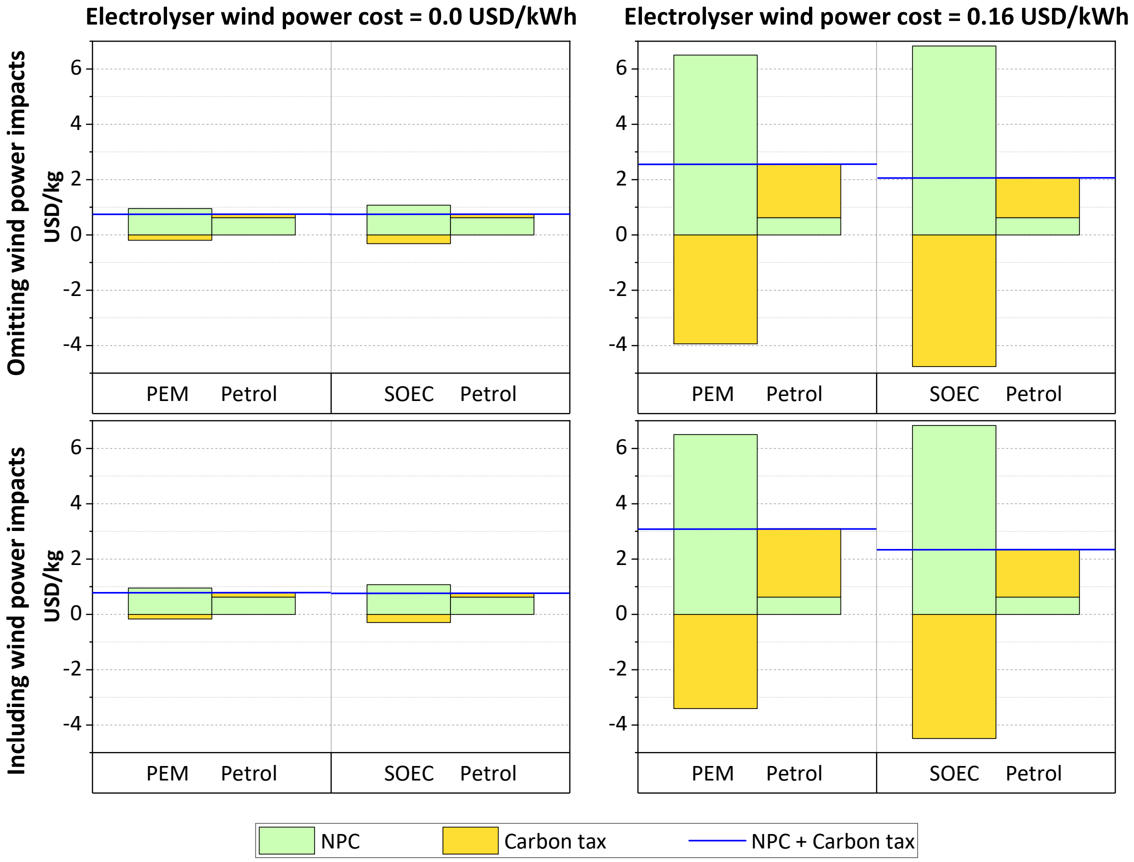


Fig. 8. Carbon tax assessment for each scenario.

3.4. *Life-cycle assessment (LCA)*

Consistent with the economic assessment, we evaluated the LCAs of the production processes for two different scenarios: (i) omitting the environmental impacts of the wind electricity powering the electrolysers; and (ii) including those impacts. The former case assumes that the required electricity is surplus electricity linked to fluctuating wind power [95,96]; consequently, the associated environmental impacts are, in this case, fully allocated to the grid electricity. The LCA results at the endpoint level are shown in Fig. 9, and at the midpoint level in Fig. 11, whereas their corresponding breakdowns are shown in Fig. 10, and in Fig. 12, Fig. 13 and Fig. 14, respectively.

3.4.1. *Analysis at the endpoint level*

As shown in Fig. 9A, when the impacts of the wind electricity are omitted, both electrofuels outperform fossil petrol in all the endpoint categories, *i.e.*, human health, resources, and ecosystems, where the gap is wider in the latter category. Hence, in this case, there is no burden-shifting, *i.e.*, no detrimental side effects linked to the shift from one technology to the other [100,101].

However, when the impact of wind power is accounted for (see Fig. 9B), both electrofuels outperform their fossil analogue in only two out of three endpoint categories, *i.e.*, ecosystems and resources, at the expense of worsening the performance in human health.

Comparing both electrofuels, it can be observed how the *SOEC* fuel has a lower environmental impact on human health and ecosystems, and a slightly higher impact in the resources category, for both scenarios. The reason for this is that, at the operating conditions assumed, the co-electrolysis-based process is more efficient due to its relatively lower consumption of feedstock (4.3 kg_{CO2}/kg_{fuel} and 6.1 kg_{H2O}/kg_{fuel} vs 4.6 kg_{CO2}/kg_{fuel} and 4.1 kg_{H2O}/kg_{fuel}, for the *PEM* and the *SOEC*-based fuels, respectively), despite displaying similar utilities consumption rates (33.5 $kWh_{electricity}/kg_{fuel}$, 4.4 $kWh_{cooling\ water}/kg_{fuel}$ and 0.17 $kWh_{propane\ refrigeration}/kg_{fuel}$ vs 34.9 $kWh_{electricity}/kg_{fuel}$, 3.6 $kWh_{cooling\ water}/kg_{fuel}$ and 0.14 $kWh_{propane\ refrigeration}/kg_{fuel}$, for the *PEM* and the *SOEC*-based fuels, respectively). Furthermore, co-electrolysis also leads to less emissions (1.8 $kg_{flue\ gas}/kg_{fuel}$ and 4.3 $kg_{wastewater}/kg_{fuel}$ vs 1.0 $kg_{flue\ gas}/kg_{fuel}$ and 2.5 $kg_{wastewater}/kg_{fuel}$, for the *PEM* and the *SOEC*-based fuels, respectively), as shown in Table 2.

Fig. 10 shows the breakdown in the endpoints, while the corresponding uncertainty intervals, given by the 2.5/97.5 percentiles, are shown in Appendix E, Fig. E-1, for both scenarios. As seen, in the human health and the ecosystems categories, the main contributor is the electricity, followed by the steel and the propane refrigeration. In contrast, in resources, the main contributor is the CO_2 followed by the electricity and the propane refrigeration. Note that natural gas, compressed air, and CO_2 lead to environmental credits in some endpoint impact categories. The main negative contribution of CO_2 is due to the carbon captured used to produce the electrofuels. At the same time, natural gas and compressed air are considered avoided products (recall that a portion of the purge gas is burnt with part of the produced oxygen from the electrolysers, as explained in section 2.4.1).

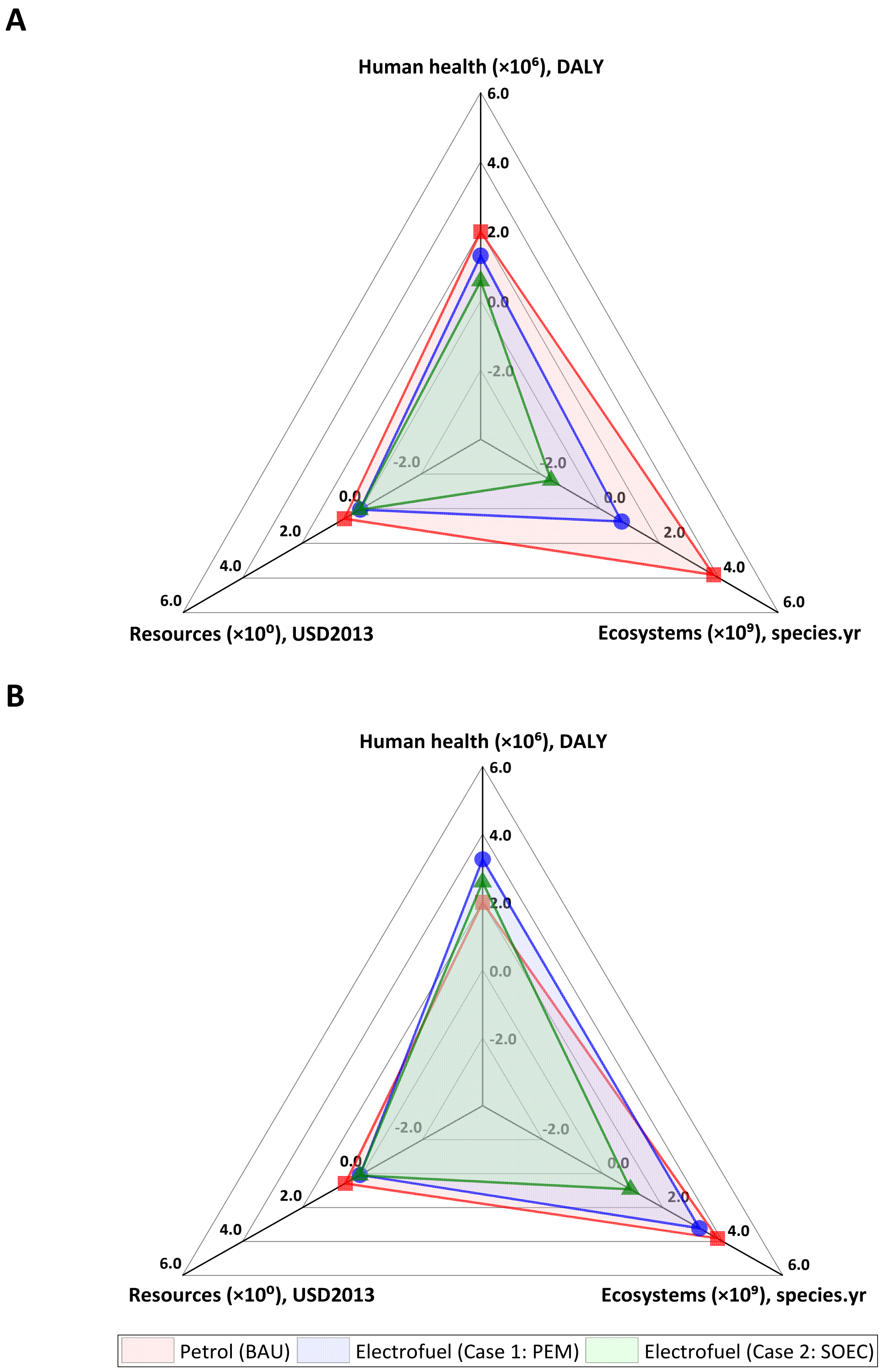


Fig. 9. ReCiPe 2016 LCA comparison at the endpoint level per kg of fuel. A: Omitting wind power environmental impacts; B: Including wind power environmental impacts

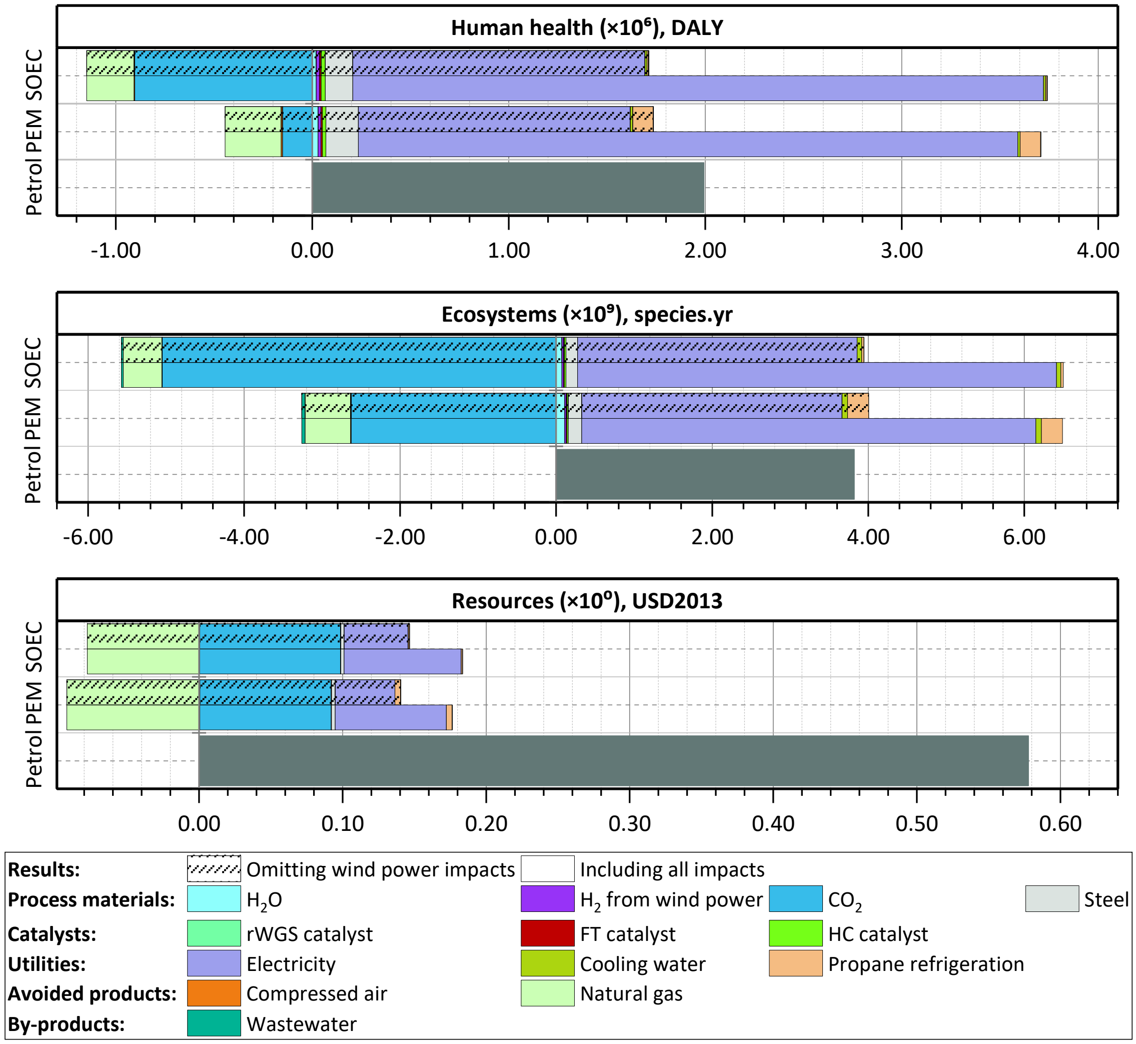


Fig. 10. ReCiPe 2016 LCA breakdown of each production process at the endpoint level per kg of fuel.

3.4.2. Analysis at midpoint level

As observed in Fig. 11, the environmental profiles of the *PEM* and the *SOEC*-based fuels at the midpoint level are close to each other for both scenarios. In both cases, omitting or not the impact of wind power, fossil petrol performs notably worse in the following three midpoint categories: global warming on human health, global warming on terrestrial ecosystems, and fossil resource scarcity. Note that the global warming indicators take negative values in both electrofuels due to the captured CO_2 and the cradle-to-gate scope that neglects the emissions during fuel combustion in vehicles.

Those negative values in the climate change midpoints, however, are counterbalanced by positive impacts in other midpoint categories, *e.g.*, fine particulate matter, ozone formation on terrestrial ecosystems, terrestrial acidification, freshwater eutrophication, human carcinogenic and non-carcinogenic toxicity, and land use. This latter finding confirms that burden-shifting also takes place in different midpoint indicators.

Analysing the breakdown of midpoints (Fig. 12, Fig. 13 and Fig. 14), we find that the environmental impacts related to the captured CO_2 and the electricity consumption are the major contributors in most midpoints. This is due to the fact that the CO_2 is allocated part of the burdens associated with electricity generation in coal power plants. Furthermore, the impact of propane refrigeration, which includes the electricity consumption of the refrigeration cycle and a propane makeup to compensate for an annual leakage of propane of around 4% [102], is quite substantial in some midpoint categories, *e.g.*, ionisation radiation and ozone formation.

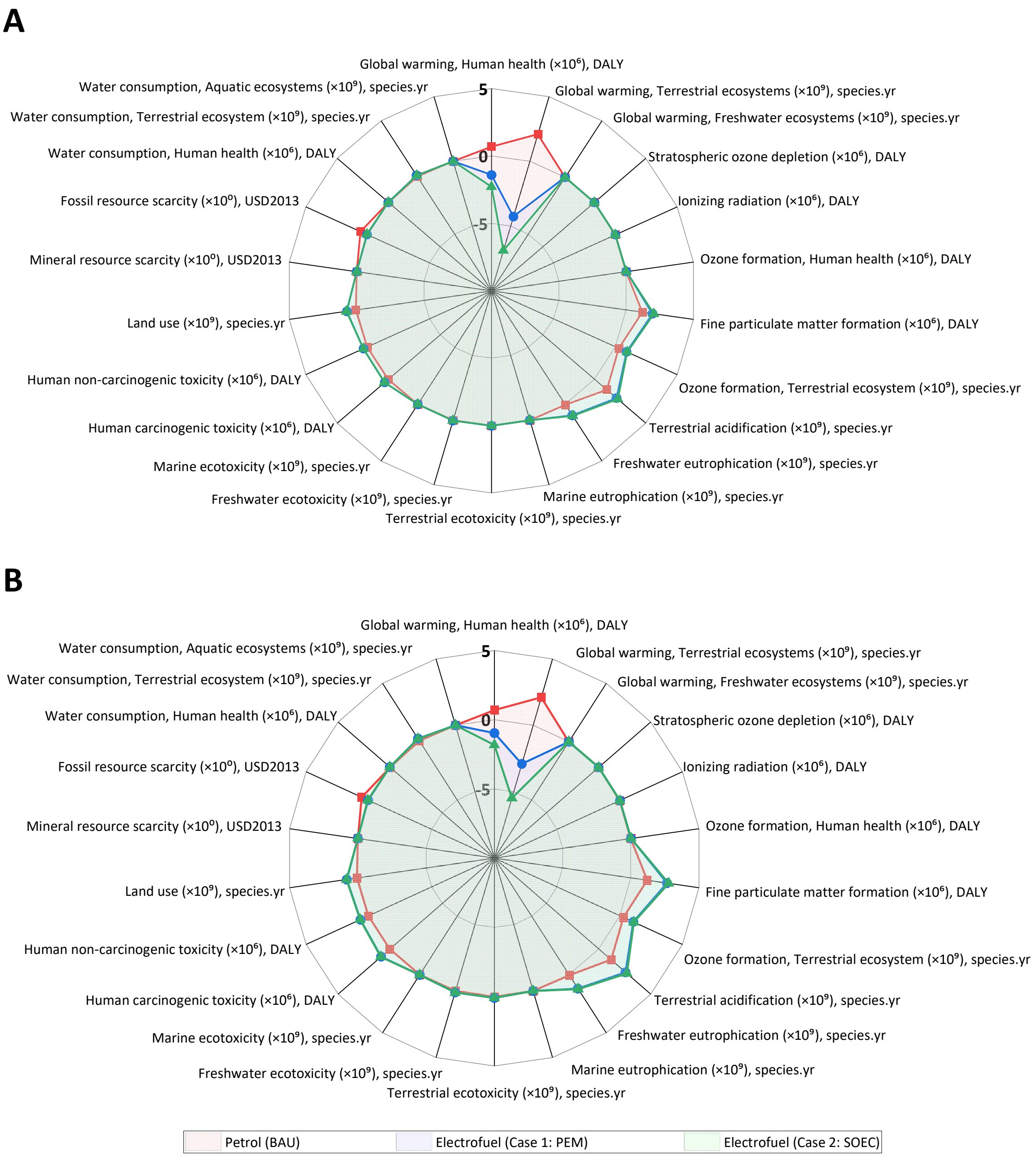


Fig. 11. ReCiPe 2016 LCA comparison at the midpoint level per kg of fuel. A: Omitting wind power environmental impacts; B: Including wind power environmental impacts

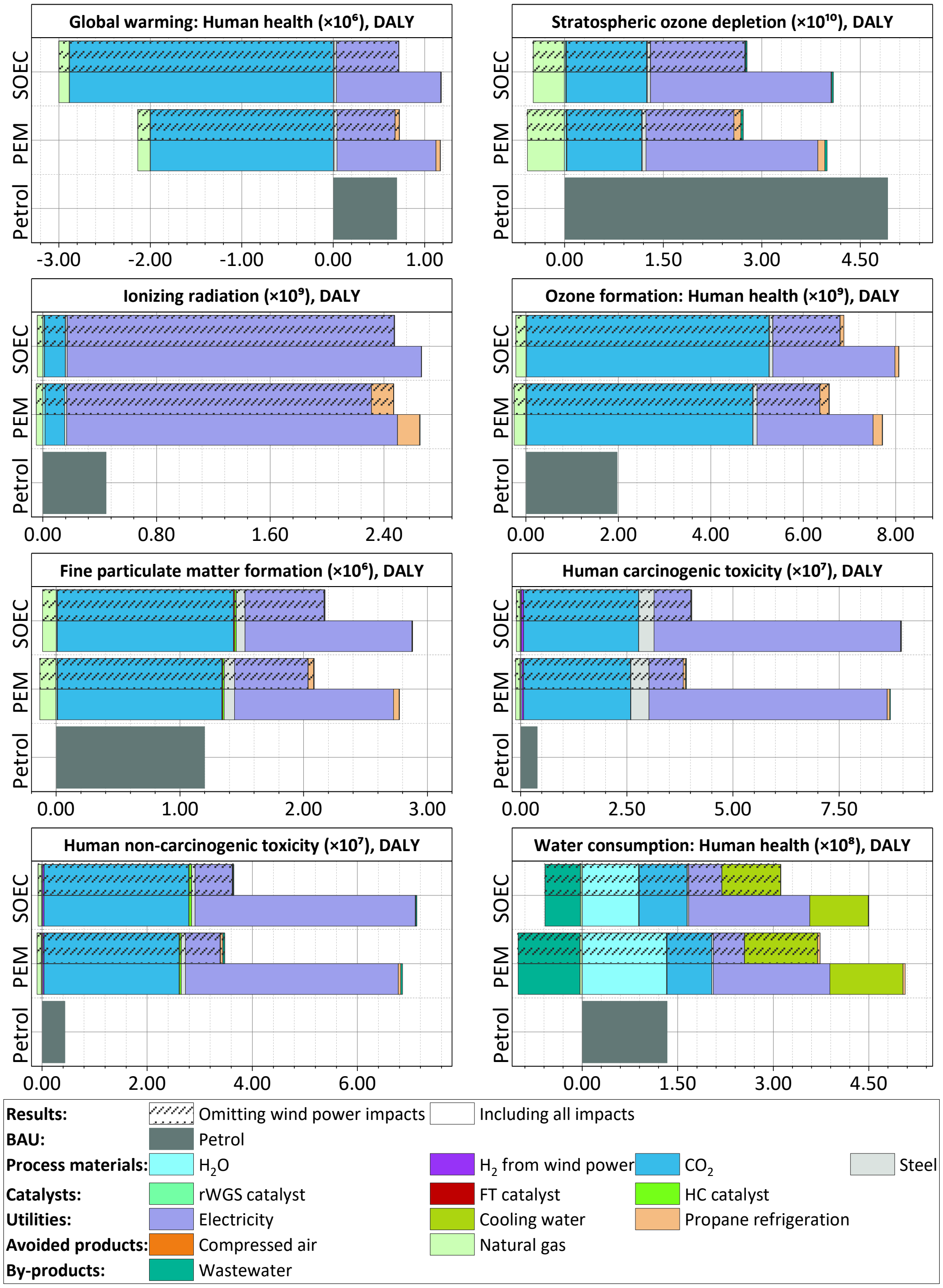


Fig. 12. ReCiPe 2016 LCA at the midpoint level of each production process; contributions to human health damage.

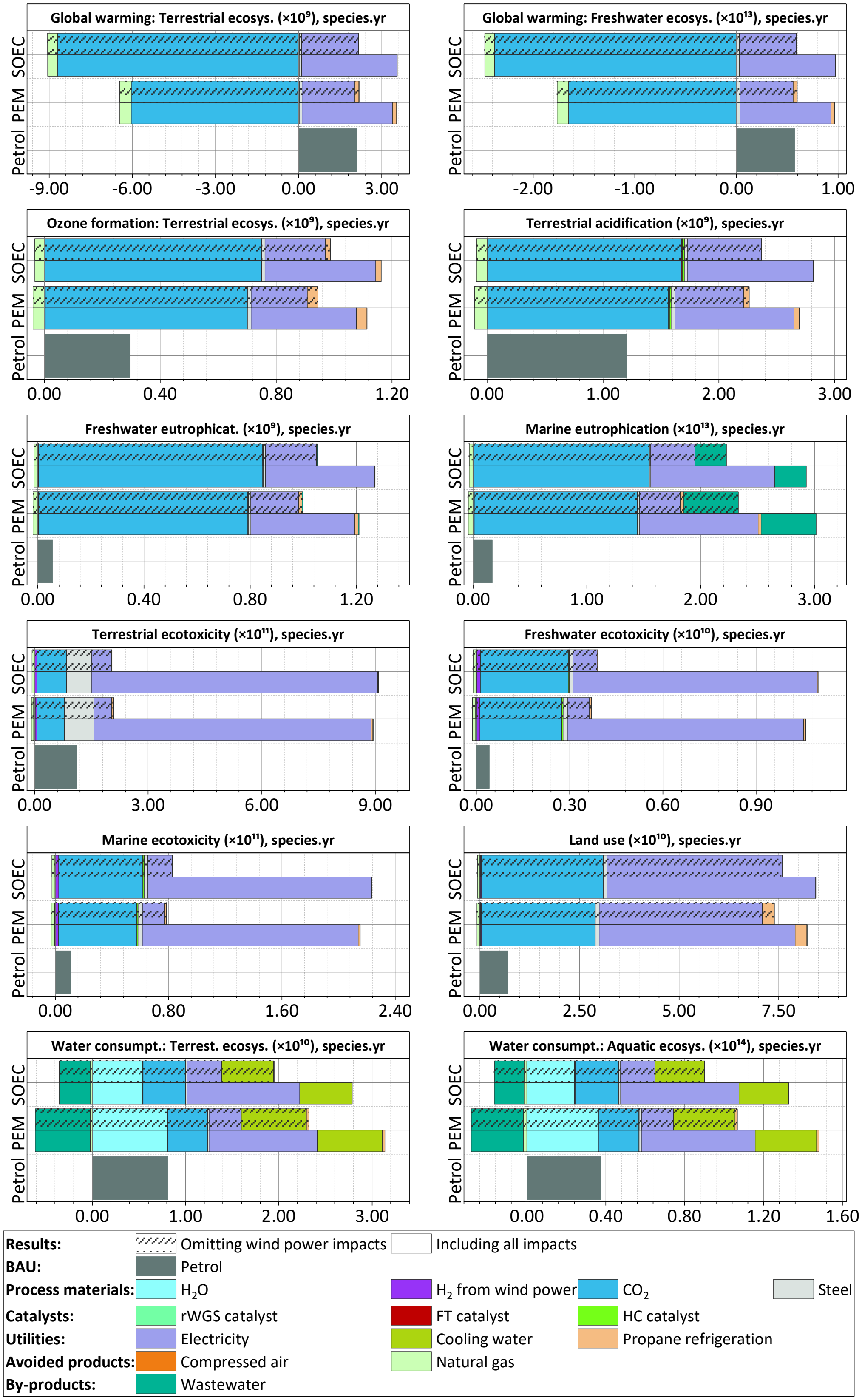


Fig. 13. ReCiPe 2016 LCA at the midpoint level of each production process; contributions to ecosystems damage.

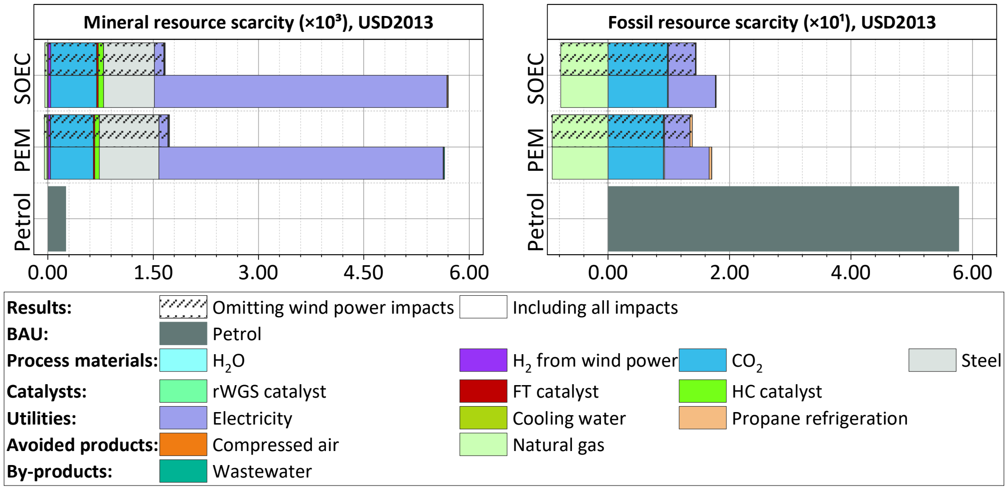


Fig. 14. ReCiPe 2016 LCA at the midpoint level of each production process; contributions to resource availability damage.

3.4.3. Comparative uncertainty analysis

As shown in Appendix E, Fig. E-1, uncertainties at the endpoint level are quite pronounced, particularly in human health and ecosystems, resulting in large confidence intervals that make it hard to discriminate between technologies. To facilitate the analysis, Fig. 15 shows the pairwise probabilities of one technology outperforming the fossil fuel in each impact category. More precisely, the figure displays the results of comparing the electrofuels with conventional petrol in each impact category across scenarios. In the figure, bars denote the percentage of samples (each corresponding to a different realisation of the uncertain parameters) in which one technology outperforms the fossil fuel. For each pairwise comparison, a total of 1000 Monte Carlo runs were simulated using Simapro v9.0, assuming a 95% confidence level.

Comparisons for water indicators are inconclusive due to the large degree of uncertainty involved, which propagates to the human health and ecosystem endpoint indicators. These results are consistent with previous studies highlighting the significant uncertainties found in the water flows reported in Ecoinvent [103]. According to the literature, an alternative should emerge as superior in at least 90% of the Monte Carlo samples [92] to attain a satisfactory level of discrimination. However, lower values would also be acceptable.

In the first scenario (omitting the impact of wind energy), the 90%-criterion is met in all the midpoints except for water consumption. In contrast, for the second scenario, the criterion is met in all the midpoints except for water consumption and stratospheric ozone depletion. Because of this, the ecosystems and human health endpoint impact categories do not meet the same criterion either. We find that in ecosystems, the *SOEC*-based fuel is better than fossil petrol with a 59.6% vs 98.8% probability, for the first scenario and the second scenario, respectively. Likewise, the *PEM*-based fuel outperforms fossil petrol with a 48.6% vs 85.1% probability, for the first and the second scenario, respectively.

Furthermore, the electrofuels outperform the *BAU* in human health, with probabilities of 35.3% vs 93.9% and 24.6% vs 78.3%, for the *SOEC* and the *PEM*-based fuels, for the first and the second scenario, respectively. Accordingly, the endpoint values are less reliable than the midpoint ones due to the highly uncertain water flows.

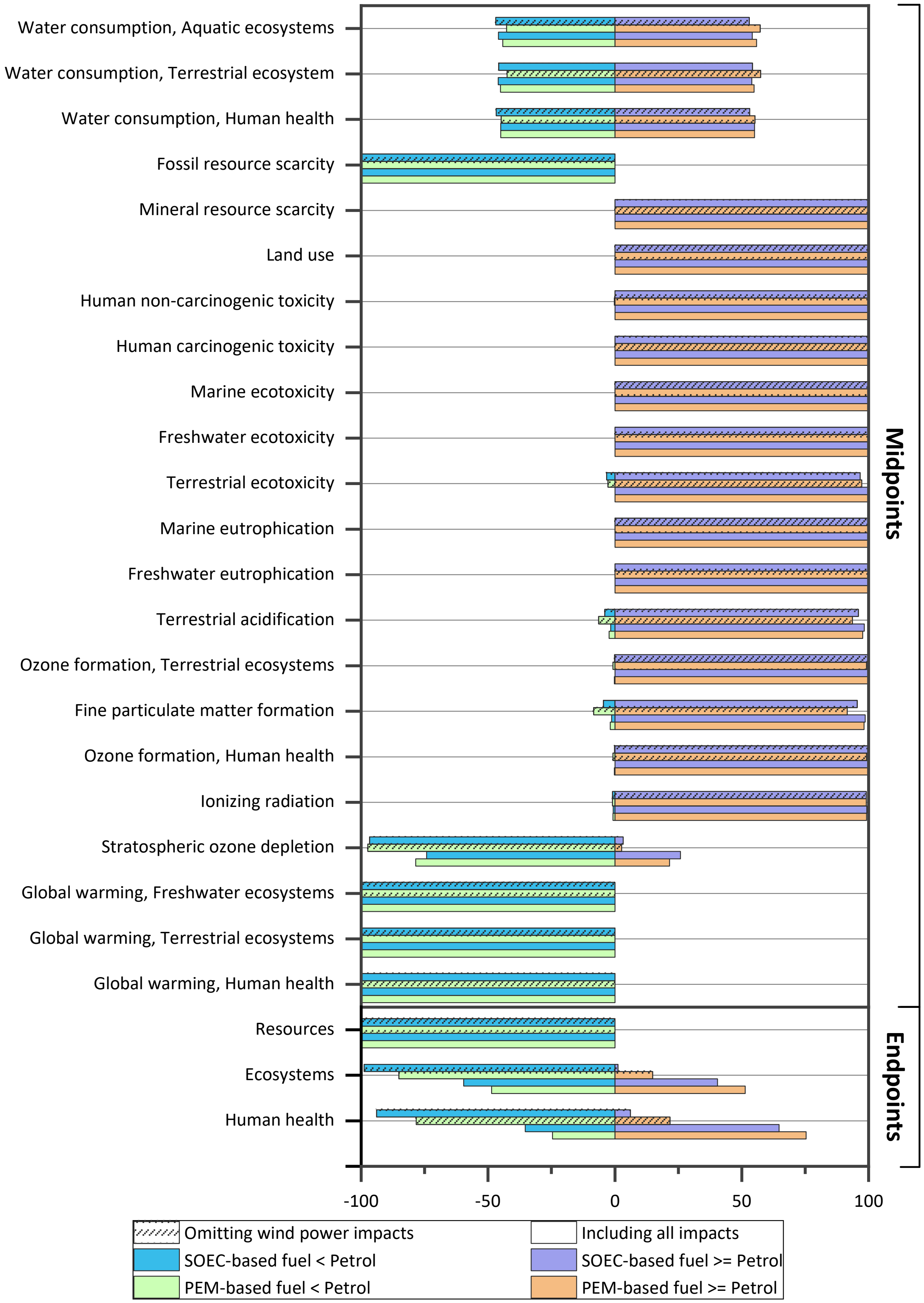


Fig. 15. ReCiPe 2016 LCA. Relative environmental uncertainty.

4. Conclusions

This study assessed the economic and environmental performance of two electrofuels produced from CO_2 and wind energy using the *PEM* and *SOEC* technologies, respectively. The total cost (*NPC*) and a range of LCA indicators based on the ReCiPe 2016 were studied considering economic and environmental uncertainties.

Our results show that electrofuels are yet economically unappealing under the current market conditions in the UK, *i.e.*, 0.16 USD/kWh$_{\text{wind electricity}}$ and 1.71 USD/gal$_{\text{fossil petrol}}$ (at least *ca.* 10.4-fold higher *NPC* than that of fossil petrol). This holds true also when considering free surplus wind energy (at least *ca.* 1.5-fold higher *NPC* than that of fossil petrol). The *SOEC*-based fuel was found to be slightly more expensive than the *PEM*-based fuel due to the higher annual production of the latter (*ca.* 6.4% to 13.0% more costly, depending on the wind electricity price). However, including externalities in the economic assessment would make electrofuels cheaper than their fossil analogue under the free wind energy scenario (petrol cost *ca.* 0.9-fold higher), but not when considering the current cost of wind electricity, *i.e.*, 0.16 USD/kWh$_{\text{wind electricity}}$.

Electrofuels could become economically appealing under a tax on CO_2 emissions as high as 212.57 USD tCO_2-eq^{-1} for the zero-cost electricity case, and 3286.45 USD tCO_2-eq^{-1} when not relaying on the excess of wind energy. The costs of wind electricity and captured CO_2 and the revenue from a potential sale of O_2 were found to be the main variables impacting the most the cost of electrofuels. Notably, the expected drop in the cost of wind electricity and captured carbon could help to make electrofuels more appealing.

According to the LCA results, when the environmental impacts of wind power are omitted, both electrofuels outperform the fossil petrol simultaneously in all the endpoint categories, *i.e.*, human health, ecosystem quality, and resources. However, when those impacts are considered, burden-shifting takes place at the endpoint level, since both electrofuels show better performance in ecosystems and resources compared to fossil petrol at the expense of worsening human health. Likewise, burden-shifting would take place at the midpoint level. The *SOEC* technology outperforms the *PEM* in human health and ecosystems but is worse in resources. This is mainly due to the fact that natural gas leads to higher environmental credits in the *PEM*-based process compared to the *SOEC*-based process. The main contributors to the total impact in both electrofuels correspond to the captured CO_2 and the required electricity.

Overall, this work points towards the need to embrace the whole range of LCA categories in the environmental assessment of alternative fuels in order to avoid the occurrence of burden shifting. It also highlights the necessity to include externalities in their assessment in order to uncover their real cost. Even considering them, subsidies would most likely be still required to make alternative fuels economically competitive, although to a lesser extent. From a more technological side, further research is required to develop more efficient and cheaper renewable energy harvesting systems and electrolysers that could help to close the economic gap with the fossil analogue.

5. Acknowledgements

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