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Drop-in and hydrogen-based biofuels for maritime transport: Country-based assessment of climate change impacts in Europe up to 2050

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ABSTRACT

Alternative fuels are crucial to decarbonize the European maritime transport, but their net climate benefits vary with the type of fuel and production country. In this study, we assess the energy potential and climate change mitigation benefits of using agricultural and forest residues in different European countries for drop-in (Fast Pyrolysis, Hydrothermal Liquefaction, and Gasification to Fischer-Tropsch fuels or Bio-Synthetic Natural Gas) and hydrogen-based biofuels (hydrogen, ammonia, and methanol) with or without carbon capture and storage (CCS). Our results show the combinations of countries and biofuel options that successfully achieve the decarbonization targets set by the FuelEU Maritime initiative for the next years, including a prospective analysis that include technological changes projected for the biofuel supply chains until 2050. With the current technologies, the largest greenhouse gas (GHG) mitigation potential per year at a European scale is obtained with bio-synthetic natural gas and hydrothermal liquefaction. Among carbon-free biofuels, ammonia currently has higher mitigation, but hydrogen can achieve a lower GHG intensity per unit of energy with the projected decarbonization of the electricity mixes until 2050. The full deployment of CCS can further accelerate the decarbonization of the maritime sector. Choosing the most suitable renewable fuels requires a regional perspective and a transition roadmap where countries coordinate actions to meet ambitious climate targets.

1. Introduction

The global maritime sector is in a transition period where critical decisions are needed to achieve a consistent decarbonization in the coming decades [1]. Without taking any measures to decarbonize this sector, the European Union's (EU) international shipping greenhouse gas (GHG) emissions are projected to grow by over 30 % between 2015 and 2050 [2]. The EU, responsible for approximately 13 % of international shipping GHG emissions [3], aims to accelerate the uptake of renewable and low-carbon fuels (RLF) in the maritime sector [2]. RLFs hold higher potential for climate change mitigation when compared to other design-, technical- and operational-related measures [4,5], and their faster implementation can importantly contribute to meeting the targets of the European Green Deal of 55 % reduction in GHG emissions by 2030 and carbon neutrality by 2050 [6]. Within the scope of the FuelEU Maritime initiative, the use of drop-in biofuels – i.e., biofuels compatible with existing engines and infrastructure – as well as new fuel alternatives (such as hydrogen-based fuels) is essential to reduce the climate impacts from fossil fuels [2].

Although shifting the fuel mix has gained a general consensus as a strategy to reduce GHG emissions in the European maritime sector, little is known about which type of RLFs are more suitable, especially considering the specificities of each country as to their different levels of access to natural resources, heterogeneous structures of production chains, and the diverse potentials of implementing environmental policies consistently throughout the next decades. Longer transport distances of feedstocks or limited access to decarbonized electricity grids, for instance, can be critical factors to consider in the selection of the most suitable technological options for fuel production. An additional challenge related to the choice of the most appropriate RLFs is the understanding of whether the projected shift in the maritime industry towards the adoption of alternative fuels such as hydrogen, ammonia, and methanol [4] will offer advantages over drop-in biofuels, given the higher risks and costs associated with changing the global maritime infrastructure in a relatively short period. From a tank-to-wake perspective, the use of zero-carbon fuels like hydrogen, for instance, is expected to better perform in terms of reduction in local pollutants such as NO_x and particulate matter, making them preferable for use at berth

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when compared to advanced biofuels, especially when considering the potential use of fuel cell technology [7,8]. On the other hand, potentially lower GHG emissions can be obtained from the production of advanced biofuels when considering a well-to-tank perspective, particularly when compared to hydrogen produced from fossil sources or even from electrolysis based on high-carbonized electricity grids [9,10]. All these factors combined rise the need for a thorough climate impact assessment of various RLF options when considering their large-scale deployment in the heterogeneous landscape of fuel production chains in Europe.

Recent studies on drop-in and hydrogen-based fuels for shipping have mostly evaluated their potentials and barriers as to their technological, safety, economic, and other relevant aspects [11–13] without any explicit comparison of their potential environmental impacts. The analyses largely focused on the energy use and costs of the different alternatives [10,14], and information on the environmental profile of biofuel technologies remains limited. Although there is an extensive literature about the emissions associated with bio-based fuels [15–18], these studies primarily describe the biodiesel performance during combustion in marine diesel engines, without consideration of the impacts from a life-cycle perspective.

Existing life cycle assessments of bio-based fuels mostly have performed analysis of individual (or a limited number of) RLFs, and each study largely differ in terms of methodological approaches, geographical locations, and system boundaries. They have assessed climate impacts of case-specific options, such as liquefied biogas from willow [19], bio-LNG from waste [20], straight vegetable oil (SVO) and biodiesel from soybean and rape [21], HTL fuels from sewage sludge [22], and a variety of other second-generation fuels from thermochemical processing of biomass residues [14,15,23]. On the other hand, life-cycle assessments (LCA) dedicated to the conversion of biomass into hydrogen, ammonia and methanol applied to the maritime transport are scarce [24,19]. Some LCA studies of bio-hydrogen production routes either have focused on cradle-to-gate impacts of different gasification process-design alternatives without any specific application [25–27] or considered the climate impacts in the road transport sector only [28,29].

Therefore, extensive comparative assessment within a common framework for quantifying the climate impacts of hydrogen-based and drop-in biofuels are missing. Moreover, there is a gap in the literature when exploring the climate change impacts of these biofuels for different countries in Europe, using the FuelEU Maritime initiative as background for the discussion. This hinders our capacity to discern the key technical factors shaping the different performances between these RLFs, how the importance of these factors varies among countries and the climate change mitigation benefits that can be expected.

This study provides a well-to-wake (WTW) climate impact assessment of both drop-in and hydrogen-based biofuels for deep-sea shipping produced from agricultural and forest residues. Drop-in biofuels include fast pyrolysis (FP), hydrothermal liquefaction (HTL), and gasification to both Fischer-Tropsch (FT) diesel and bio-synthetic natural gas (BSNG), while hydrogen-based biofuels are produced by converting residues into hydrogen, ammonia, and methanol. The analysis estimates the energy and climate change mitigation potentials of a large-scale implementation of diverse production pathways in different European countries, considering local constraints in terms of residue biomass availability and access to electricity grids with different levels of GHG intensity. As the FuelEU Maritime initiative projects that most of the feedstock used in biofuel production by 2030 come from biomass waste flows [2], biofuels are assumed to be produced from forest and agricultural residues available in Europe. This aims to minimize pressure on land resources and promote a circular economy perspective. Considering the need for faster decarbonization in the maritime industry, we include an additional pathway of biofuel production coupled with carbon capture and storage (BECCS). This can quantify the potential to deliver negative-emission biofuels to the European maritime transport sector.

Besides well-mixed greenhouse gases (CO_2, CH_4, and N_2O), near-term climate forcers (NTCFs) – i.e., short-living species in the atmosphere such as sulfur oxides (SOx), nitrogen oxides (NOx), particulate material (PM), black carbon (BC) and others – and three different climate metrics are included in the analysis to better describe the climate system response in a short-, medium-, and long-term perspective. Taking into account that the impact of choices we make today (such as the construction of new biofuel plants, changes in the engine technology, and adaptation of distribution and fueling infrastructures) will take a few years before they are implemented and they will last for decades, we carry out a prospective life cycle assessment [30] to explore how the implementation of alternative climate change mitigation policies up to 2050 will affect the climate impacts of the investigated biofuels. These policies include changes in the background system to follow certain future pathways of progressive decarbonization trends in energy, transport, material production, and other socio-economic factors as described by Integrated Assessment Models (IAMs). We test the robustness of our results with a thorough Monte-Carlo analysis that accounts for uncertainty and variability in key factors, such as transport distances of residues, biofuel conversion yields, GHG intensity of national electricity grids, industrial and distribution losses, emission factors, fuel consumption, and emission metrics.

This study is structured as follows. The methodology is described in Section 2, where the main assumptions and source of data used in the life cycle climate impact assessment of biofuels are described (and made available in the Supplementary Material). In Section 3, we present the comparisons of the climate change effects of drop-in and hydrogen-based biofuels, showing their potentials for climate mitigation for each European country and at an aggregated continental level. Finally, Section 4 discusses the main implications for each biofuel-country combination to achieve the climate mitigation targets stated by the FuelEU Maritime initiative until 2050.

2. Methods

In Section 2.1, we define the system boundaries of our life cycle assessment. The detailed description of the data used to quantify biomass residues availability in Europe and the overview of the Policy context of European maritime transport emissions are presented in Sections 2.2 and 2.3, respectively. The explanation of the technologies and assumptions made for compiling the foreground life cycle inventories and to estimate the emissions from the use of biofuels are found in Sections 2.4 and 2.5, respectively. We provide a clear description of the steps involved in the creation of future inventory databases and the uncertainty analysis in Sections 2.6 and 2.7, respectively.

2.1. Scope of the study

This study is based on a well-to-wake (WTW) climate impact assessment that embeds all inputs and emissions involved in raw material extraction, agricultural and forestry residues transport to the industrial plant, biomass conversion, biofuel distribution, and combustion (as depicted in Fig. 1). Our analysis uses a multi-metric approach [31] for the climate impact analysis, with three complementary climate metrics to capture different dimensions of the climate system response [32]: in the short-term with the 20-year global warming potential (GWP20), in the medium-term with the 100-year global warming potential (GWP100), and the long-term with the 100-year global temperature potential (GTP100). Previous studies highlighted the importance and temporal variability of the climate impacts of NTCFs from shipping [5,33,34], and a multi-metric approach can duly inform about the different timescales of the climate impact evolution.

As described in the further subsections, the critical data used in study are based on previous studies available from the literature and own calculations. The different data sources and resolution can be found in Table S1 of the supplementary material.

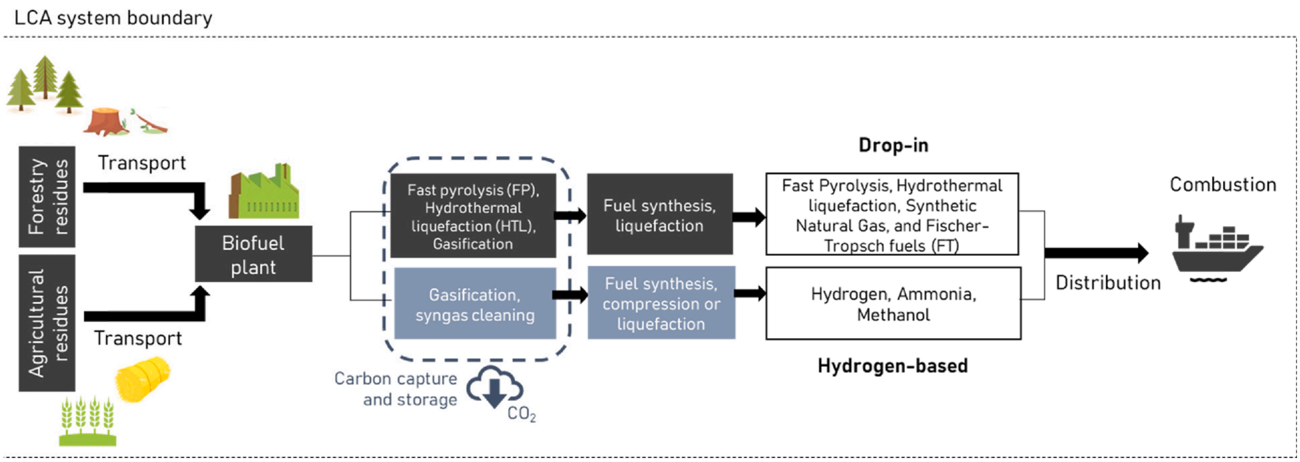


Fig. 1. Simplified scheme and system boundaries of our study. The figure shows the two biofuel types considered for deep-sea maritime applications: drop-in and hydrogen-based biofuels. A full description of biomass collection and transport, biofuel production technologies and emissions from combustion can be found in the Sections 2.2, 2.4 and 2.5, respectively.

2.2. Biomass resource availability and life-cycle inventory

Our study explores the potential biofuel production from agricultural and forest residues in European Union Member States (Austria, Belgium, Bulgaria, Croatia, Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain, and Sweden), Norway, Switzerland, and the United Kingdom. Data on crop residues are based on a spatially explicit assessment that quantified the potential agricultural residues (i.e., straw, stem, leaves, chaff, and stubble) available in Europe at a 1 km grid resolution [35]. The analysis included the main crops cultivated in Europe, namely wheat, rye, barley, oats, maize, rice, rapeseed, and sunflower. Residue potentials are provided at different removal rates (theoretical, technical, environmental, and sustainable). For biofuel production, we consider the country-specific residue availability according to the 'sustainable' potential, which represents a sustainable residue removal rate that avoids the depletion of soil organic carbon stocks in the cultivated areas. Data on forestry residues – i.e., branches and harvest losses – were taken from a study that mapped the spatial distribution of woody biomass potentials in Europe at a 10 km grid resolution level [36]. From the different biomass potentials estimated, we consider the 'base' potential that refers to forest residues availability in line with current guidelines of sustainable forest management and covers legal restrictions from management plans in protected areas. The estimates of biomass residue potentials used in our study are thus conservative.

The resulting sustainable potential of crop and forestry residues in Europe is 123.5 and 36.0 M_{tondb} yr^{-1}, respectively. Gridded data were aggregated per country (see Table S2 in the Supplementary Material) to facilitate integration with other inventory inputs, such as electricity from the grid, provided at a country level [37]. Although a portion of the crop and forest residues are currently in use by other industries [38,39] and other sectors (e.g., aviation) may exacerbate competition for biomass residues, this study investigates the total potentials for the maritime transport sector and thus considers that all available residues will be converted into biofuels. We assume no life-cycle greenhouse gas emissions to produce agricultural and forest residues since the entire environmental burdens of biomass production were allocated to their main products, such as grains, seeds, and wood. Therefore, we only account for the life-cycle impacts associated with the collection and transport of residues from the field to the biofuel plant. The average residue transport distances per country consider the residue availability per area in different European countries and the optimal distances from the field to biofuel plants of a capacity of 560 thousand tons of dry biomass per year. The transport from the field to the industrial plant is based on residues' moisture content ranging from 15 to 40 % [35].

2.3. Policy context of European maritime transport emissions

In this study, we refer to the European Parliament's Regulation EU 2017/352 which establishes that the GHG intensity of the energy used on-board by a ship must gradually decrease below specific limits over the next decades. Based on the weighted average of emission factors of current fossil fuel use in the European Economic Area (EEA), the well-to-wake life cycle GHG intensity of fossil fuels is defined as 87 $gCO_{2\text{-eq}}$ MJ^{-1}. The percentage reductions achieved by RLFs in relation to this factor should be at least −7% in 2030, −26 % in 2040, and −75 % in 2050 under Policy Option 3 (PO3), whose projection fosters over-achievement and encourage the development of more advanced, zero-emissions technologies [2]. The methodology considers a well-to-wake perspective, with the GHG intensity reduction targets based on GWP100 without the inclusion of NTCFs.

The Annual Report on CO_2 Emissions from the EU Maritime Transport [40] reports the use of ca. 44 million tons of fossil fuels per year, namely heavy fuel oils (HFO), marine gas oil (MGO), and liquefied natural gas (LNG). Based on their consumption and average lower hearting values [41], the current energy demand from the EU maritime sector is estimated at ca. 1826 PJ or 43.6 Mtoe yr^{-1}. The official annual emissions correspond to 138 $MtCO_{2eq}$ [40], but this estimate only considers emissions from combustion in the ships and does not include life-cycle emissions associated with fossil fuel production. To be consistent with the total well-to-wake (WTW) perspective considered in this study, we re-estimate the EU maritime transport's emissions in the results section. In the results section, we also refer to the 2030 Climate Target Plan [6], which aims at both reducing at least 55 % GHG emissions below 1990 levels by 2030 and reaching climate neutrality by 2050. The assumptions made for the calculations are detailed in the Tables S3-S6 in the Supplementary Material.

2.4. Biofuel pathways

We provide a comparison between drop-in and hydrogen-based biofuel pathways produced from biomass residues in Europe to identify the potential advantages and disadvantages of a transition towards a different maritime transport infrastructure. Four drop-in marine biofuel production pathways are considered: bio-synthetic natural gas (BSNG), fast pyrolysis (FP), hydrothermal liquefaction (HTL), and gasification with Fischer-Tropsch synthesis (FT). Their climate footprint in Norway has been presented in detail in a recent paper [34]. The main advantage of drop-in biofuels is the interchangeability with current fossil fuels as nearly no adaptation in the existing ships' engines technology is required and the existing bunkering infrastructure can be used [42]. Moreover, we included three additional pathways for drop-in biofuel production with carbon capture and storage systems (BECCS) to explore their potential for delivering negative-emission fuels to the maritime transport sector in Europe: hydrothermal liquefaction with CCS (HTLCCS), bio-synthetic natural gas with CCS (BSNGCCS), and Fischer-Tropsch synthesis with CCS (FTCCS). Fast pyrolysis integration with carbon capture and storage was not considered because of its low feasibility and lack of studies on mass and energy balances describing such integration.

The hydrogen-based fuels are bio-ammonia (BNH3), bio-hydrogen (BH2), and bio-methanol (BMEOH), which are considered the most promising alternatives to decarbonize the maritime sector [4,43]. Hydrogen is a carbon-free fuel with very low emission of pollutants during its combustion. Converting hydrogen into other energy carriers, such as ammonia or methanol, results in fuels that can be more easily transported and stored, and which are also more compatible with existing infrastructure or end-use technologies [43]. They still depend on further developments in the distribution and bunkering infrastructure for deep-sea applications, but current estimates project internal combustion engines (ICE) or fuel cell technologies to be available for onboard use in the next five or ten years [4]. As for drop-in biofuels, we further model three additional scenarios coupled with carbon capture and storage systems (BNH3CCS, BH2CCS, and BMEOHCCS).

2.4.1. *Drop-in biofuels*

This subsection introduces the main technologies assumed to produce drop-in biofuels. It provides a compilation of thermochemical pathways whose outputs can substitute heavy fuel oil, marine gasoil and liquified natural gas.

2.4.1.1. Fast pyrolysis. In the pathway of Fast Pyrolysis (FP), biomass residues are rapidly heated, and the pyrolysis vapors are condensed to produce fast pyrolysis bio-oil according to unit operations described in Tews and Elliott (2014). Considering the high oxygen content of FP bio-oil and its relatively low heating value (LHV), an additional stabilization step via the addition of hydrogen is required [45]. In this study, we assume that hydrogen is produced through alkaline electrolysis at an electricity use of approximately 3.8 kWh/Nm^3 H_2 [46]. The resulting output is a stabilized bio-oil with a higher LHV that is compatible to displace fossil heavy fuel oil (HFO) in deep-sea shipping (see Tables S7-S8 in the Supplementary Material).

2.4.1.2. Hydrothermal liquefaction. For hydrothermal liquefaction (HTL), we base our inventory on material and energy balances from a previous study [47]. The biocrude oil obtained from supercritical hydrothermal liquefaction of biomass residues undergoes upgrading through hydrotreatment – similarly to FP, where hydrogen is produced from alkaline electrolyzers at the biofuel plant. The outputs of HTL are drop-in renewable marine fuel (residue fraction) and diesel (distillate fraction) – which can displace HFO and MGO (marine gasoil), respectively. Since we have co-products in this biofuel plant, we distribute the climate impacts of each fuel output based on the energy allocation method. The integration of HTL with carbon capture and storage systems (HTLCCS) is based on mass and energy balances [48] which describes a Selexol^{TM} process adapted to absorb and separate CO_2 in an industrial plant with a process design similar to HTL without CCS [47]. In this configuration, CO_2 from both the effluent gas from HTL and the hydrotreater effluent gas are captured. In the HTLCCS pathway, approximately 21 % of the carbon in the biomass is captured and stored [48]. Compared to other pathways assessed in the study, such a fraction can be considered relatively low because HTL converts more biomass into liquid biofuels and, consequently, a lower share of the carbon input is transformed into gaseous byproducts. All biofuel pathways with CCS in this study assume the additional material and energy used to transport the CO_2 over a 200 km pipeline and store it in a saline aquifer based on a previous study [28] (see Tables S9-S13 in the Supplementary Material).

2.4.1.3. Gasification with Fischer-Tropsch synthesis. Gasification with Fischer-Tropsch (FT) synthesis is based on a previous study [49], which describes the gasification plant that converts dried lignocellulosic material into syngas – which consists primarily of carbon monoxide and hydrogen – that, in turn, undergoes a catalytic reaction that produces diesel. Considering that FT also generates gasoline and surplus electricity, the distribution of environmental impacts to co-products is based on energy allocation. In this assessment, we add a pathway that integrates the FT biofuel plant [49] with a CCS system capturing the carbon-dioxide-rich streams after syngas cleaning (acid gas removal step), which corresponds to ca. 49 % of the carbon in the biomass. Additional materials and energy used in the absorptive unit based on monoethanolamine (MEA) and in the CO_2 drying and compression sections were included according to a previous study [50]. Because of the electricity penalty associated with CCS in the biofuel plant (ca. 3.7 % in the overall energy efficiency), the FTCCS pathway turns out to be a net importer of electricity (see Tables S14-S17 in the Supplementary Material).

2.4.1.4. Gasification of bio-synthetic natural gas. The production of bio-synthetic natural gas (BSNG) is based on the gasification of biomass residues according to previous studies with mass and energy balances [51,52]. Once syngas is cleaned and upgraded, it undergoes a methanation step where the BSNG – ca. 97 % methane – is dried, liquefied, and stored at the biofuel plant. Complimentary data used for the estimation of chemical inputs consumed in the plant with a similar process design [53,54]. We also considered methane losses in the biofuel plant during the gas upgrading step varying from 0.04 % [53] to 0.7 % [51]. The mass and energy balances for the BSNG pathway integrated with CCS are based on a recent study [55], which consider a biofuel plant with the same process design [51]. In the BSNGCCS pathway, carbon dioxide separated after an MEA-based acid gas removal section is then captured in the biofuel plant and corresponds to ca. 32 % of the biomass carbon input (see Tables S18-S21 in the Supplementary Material).

2.4.2. *Hydrogen-based biofuels*

Considering the possibility of deeper changes in the maritime fuel supply chain structure, this subsection presents the main technologies associated with the production of non-drop in biomass derived fuels such as hydrogen, ammonia, and methanol.

2.4.2.1. Bio-hydrogen production. For hydrogen production (BH2), lignocellulosic biomass is milled and dried. Thereafter, it undergoes an indirect gasification process as described by a previous study [56], which provides data on energy and mass balances that we used to compile the foreground inventory. A syngas cleaning step removes fine particles and sulfur compounds, and then a water gas shift (WGS) process makes the carbon monoxide within the syngas react with water to produce hydrogen and carbon dioxide. The produced hydrogen is then separated from the rest of the compounds (CO_2, CO, CH_4, and others) in a pressure swing adsorption (PSA) unit. In this study, we split the BH2 pathway into two sub-pathways to improve the discussion of the impacts of compressed and liquid hydrogen distribution in Section 3.3. The electricity consumption involved in the process of hydrogen compression before storage can vary between 1.7 and 6.4 kWh per kg H_2 when considering actual compression energies and compressor inefficiencies [57]. For the BH2-P (which stands for pressurized bio-hydrogen) pathway, we assumed an average of 4.05 kWh per kg H_2 aiming at a final pressure of 350 bar. On the other hand, to reduce the physical size of the ships, liquid hydrogen is an option because the volume and pressure requirements are much smaller than those associated with the storage of compressed hydrogen gas. For liquid bio-hydrogen (BH2-L), the main advantage is the storage at ambient pressures, however, more electricity is demanded (from 10 to 13 kWh per kg H_2) to reach the cryogenic state (temperature of −253 °C) [57]; we assumed an average of 11.5 kWh per kg H_2. In the BH2-L pathway, we also consider that the liquid hydrogen incurs other impacts such as boil-off (hydrogen evaporation) and transfer losses of up to 25 % [58].

The integration of bio-hydrogen production with CCS (BH2CCS) follows the description from a previous study [27], whose process has a similar configuration to the plant without CCS [56]. In BH2CCS, the gaseous compounds separated after the PSA unit are combusted in a boiler to produce the steam and electricity required in the plant. Thereafter, the exhaust gas undergoes a gas separation polymeric membrane process for CO_2 capture, which can take up to 51 % of carbon input in the biomass. Carbon dioxide is then compressed and sent out to the biofuel plant using pipelines [28] (see S22-S25 in the Supplementary Material).

2.4.2.2. Bio-ammonia production. The production of bio-ammonia (BNH3) is based on [59] and [61], which consider entrained-flow gasification of biomass. Syngas is cleaned and undergoes a conditioning process through partial air oxidation. The resulting syngas is rich in H_2, and it is compressed and combined with nitrogen from the air to produce ammonia through the Haber-Bosch process. Since ammonia is produced at between 100 and 250 bar, there is not any additional electricity consumption to achieve higher pressures for compressed ammonia distribution. Considering that the energy required to liquefy ammonia is relatively low – ca. 0.2 kWh per kg NH_3 [61] –, instead of splitting BNH3 into compressed and liquid pathways, we just included the additional energy consumption to obtain liquefied ammonia as an additional parameter in the uncertainty analysis.

The integration of BNH3 with CCS is based on the capture of carbon dioxide via physical absorption after the syngas cleaning step, as previously described by previous study [62]. Based on the original bio-ammonia production process [60], we estimate an amine-based adsorptive capture process taking 90 % of the carbon released after syngas purification, which results in a 58 % uptake of the biomass input carbon in the BNH3CCS pathway. The additional materials and energy used both in the MEA-based absorptive unit and CO_2 drying and compression sections were based on data from previous study [50] (see Tables S26-S29 in the Supplementary Material).

2.4.2.3. Bio-methanol production. Mass and energy balances of gasification followed by bio-methanol synthesis (BMEOH) are based on data from a previous study [63]. In this process, carbon monoxide and hydrogen within syngas react in the presence of catalysts – usually based on copper oxide, zinc oxide, or chromium oxide – to produce methanol. We based our inventories on the high-pressure case scenario, whose outputs are methanol and surplus electricity. Energy allocation was used to distribute the impacts between co-products. The same report [63] describes the integration of methanol with a carbon capture system (BMEOHCCS), based on the compression of carbon dioxide separated via the Rectisol^{TM} process at the acid gas removal stage. There is an electricity penalty of ca. 2 % in the overall energy efficiency of the plant because of consumption in the CO_2 compression unit. This results in a negative electricity balance; consequently, additional electricity is purchased from the grid. BMEOHCCS pathway captures approximately 52 % of the biomass carbon input (see Tables S30-S33 in the Supplementary Material).

2.5. Emissions from marine fuels combustion

For emissions of drop-in biofuels and bio-methanol combustion in ships, we follow a previous method [34] that adapts emission factors from fossil fuels [41] to biofuels using data from a previous study [23] and by considering combustion tests comparing emissions from fossil and biofuels in internal combustion engines [64]. The emission factors considered in this study are related to carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), carbon monoxide (CO), sulfur oxides (SO_x), nitrogen oxides (NO_x), particulate material (PM_{10} and $PM_{2.5}$), non-methane volatile compounds (NMVOCs), and black carbon (BC).

Although marine fuel cells are expected to be integrated into power systems over the next years, the first hydrogen- and ammonia-fueled vessels running with internal combustion engines will soon be entering the world fleet [4]. Hydrogen and ammonia offer zero-emission combustion in terms of CO_2, CO, and hydrocarbons. On the other hand, in the presence of air and high temperatures, their combustion results in NO_x emissions. For that reason, we consider nitrous oxide emission factors of hydrogen and ammonia combustion based on combustion data from previous studies [65,66]. In the case of ammonia, eventual emissions from pilot fuel used to improve combustion were not considered since such impacts could be strongly minimized by using drop-in biofuels instead of fossil diesel in the future (see Table S34 in the Supplementary Material).

2.6. Prospective life-cycle assessment

We perform a prospective LCA that considers the influence of future technological evolution on background systems over the next three decades. Using premise version 1.0.8 [30], we align life cycle inventories of key processes in ecoinvent 3.8 [67] with the outputs of the REMIND Integrated Assessment Model [68]. The output databases embed technological improvements in electricity production mixes, power plant efficiencies, average fleet, and energy mix used for transport, and advanced technologies to produce hydrogen, clinker, cement, and metals, among others. New background inventories are built to represent the technological scenarios for 2030, 2040, and 2050 according to the ‘SSP2 - Middle of the Road’ where the world faces intermediate challenges for climate change mitigation and adaptation, with reasonable population growth, lower energy use, but slow progress in achieving sustainable development goals [69]. In terms of environmental policy scenario, we consider the National implemented Policies scenario (NPi) which describes energy, climate, and economic projections for the period until 2030, based on currently implemented national policies for achieving the internationally pledged INDC (Intended Nationally Determined Contributions) targets stipulated after the Paris Agreement. In practice, NPi represents a scenario in which climate action up to 2050 is not becoming more stringent than that implied by currently implemented policies [70].

The emissions associated with current electricity mixes at the country level are based on the ecoinvent 3.8 database [67]. As REMIND projects improvements in European electricity production as an aggregated region of the world, we assume that a similar pace of decarbonization – in terms of percentage reductions – will be achieved at the country level over the next decades (2030–2050) (see Table S35 in the Supplementary Material).

2.7. Uncertainty analysis

A Monte-Carlo analysis is performed to test the robustness of our analysis to a range of uncertainty factors: residue’s transport distances, lorry size, residues moisture, biofuel conversion efficiencies, GHG intensity of the European electricity grid representing the average emissions of different countries [67], distance from biofuel plant to seaport, methane leakage rate (for BSNG), liquid hydrogen distribution losses, emission factors from fuel combustion, and emission metrics for NTCFs (see Table S34 in the Supplementary Material). For biomass transport, both the average residue availability per area in different European countries and the optimal distances from the field to biofuel plants are considered. For biofuel conversion efficiencies, we base our ranges on the literature for all pathways: FP [44,45], HTL [44,47], BSNG [51,53], FT [49,63], BH2 [27,29,71], BNH3 [60,72,73], and BMEOH [63]. Uncertainties associated with NTCFs are based on data from a previous study [32]; for emission factors from combustion, they are a combination of uncertainties on engine fuel consumption and specific emissions of gases as reported by [41]. In the Monte-Carlo analysis, parameters were assumed to follow a triangular distribution, and results are produced for 10,000 individual simulations. In the results we show both a deterministic value produced by using the average factor of each source of uncertainty and the median, 5th and 95th percentile confidence bounds from the Monte Carlo analysis.

3. Results

In Sections 3.1 and 3.2, drop-in and hydrogen-based biofuels are compared under different climate metrics representing short-, medium-, and long-term climate impacts. In Section 3.3, the FuelEU Maritime decarbonization goals are considered as background for discussion of the climate impacts of different biofuel technologies both at a country and European level.

3.1. Drop-in biofuels

The climate impacts of drop-in marine biofuels and their associated uncertainties are shown in Fig. 2, which considers Europe as an

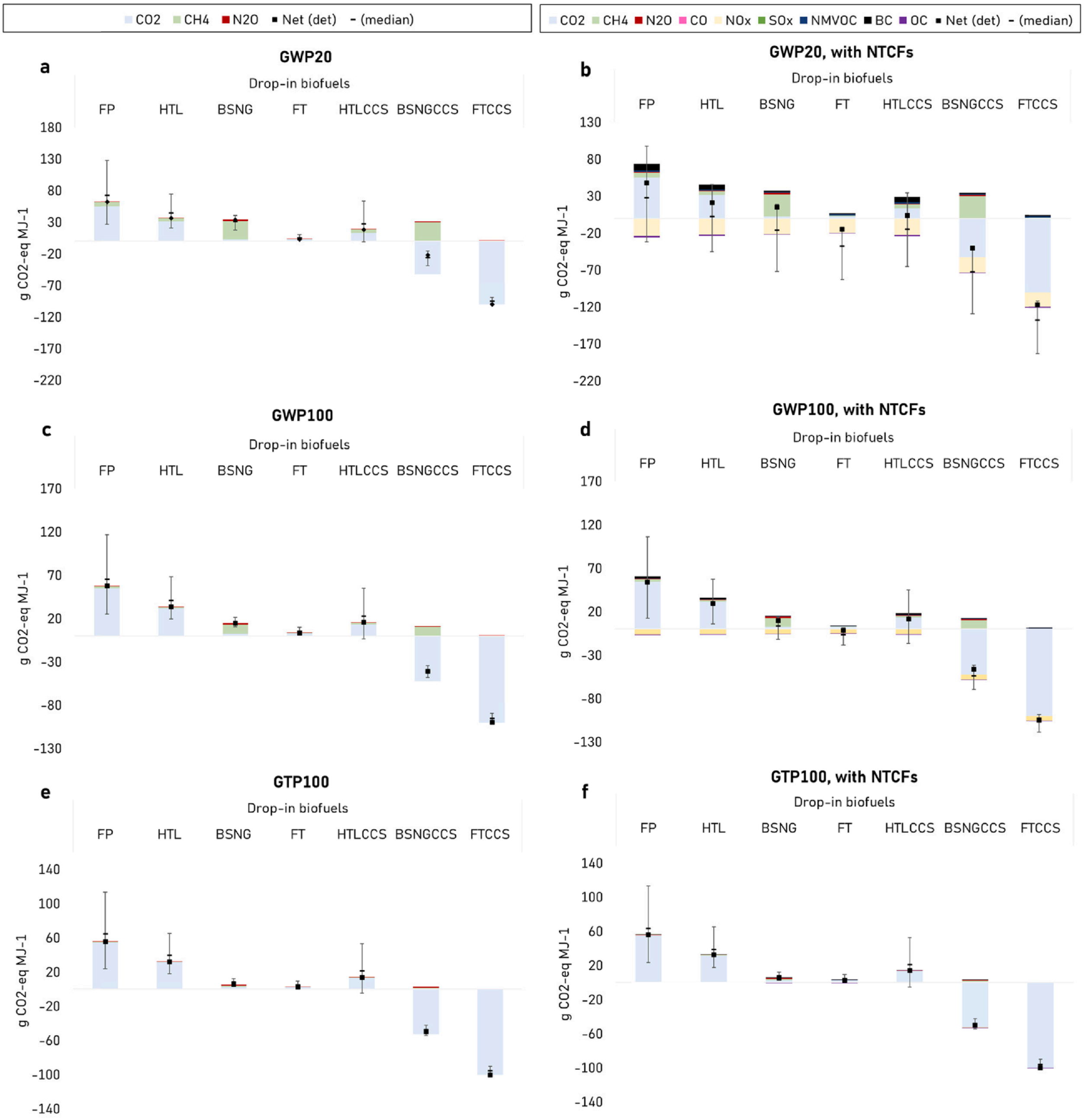


Fig. 2. Climate change impacts of drop-in biofuels from agricultural and forestry residues in Europe under multiple climate metrics with or without contributions of near-term climate forcers. Results are shown for GWP20 (a, b), GWP100 (c, d), and GTP100 (e, f). The biofuel pathways are the following: Fast Pyrolysis (FP), Hydrothermal Liquefaction (HTL), gasification to Bio-synthetic natural gas (BSNG), and Fischer-Tropsch (FT) without and with carbon capture and storage systems (HTLCCS, BSNGCCS, FTCCS). Breakdown and net impact (square-shaped point) refer to the deterministic analysis (det); the whiskers and lines show median, 5th, and 95th percentile confidence bounds from the Monte Carlo analysis, with variation in y-axis scales in the different panels.

aggregated region of the world. Among the pathways without CCS, biofuels produced from gasification – BSNG and FT – have the lowest climate impacts. In the medium term (GWP 100), the medians of BSNG and FT are 16 and 5 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively, with relatively small uncertainty ranges because the biofuel plants do not depend on the electricity from the grid, which is the dominant factor in the uncertainty analysis given the large difference in the carbon intensity among European countries (from 19 to 976 $gCO_{2\text{-eq}}$ kWh^{-1}). For the BSNG pathway, the largest share of the climate impacts come from methane slip rates in the combustion stage, which are assumed to vary from 0.2 to 2.5 g CH_4/kWh depending on the engine type used in deep-sea shipping [41]. FT has the lowest impact among all pathways without CCS and most of the emissions depend on the collection of biomass residues which, in turn, are affected by the uncertainty in transport distances across the different European countries. On the other hand, FP and HTL have the highest medians – 65 and 40 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively – with most of the emissions (93 % and 81 %, respectively) associated with industrial electricity consumption. Since hydrogen is provided by electrolysis in the biofuel plant – instead of natural gas for steam methane reform (SMR) – the variability of the European electricity grid becomes the largest driver of uncertainty. When considering the range of life cycle GHG emissions in Europe, medium-term climate impacts of FP and HTL vary between 25 and 117 and 19–68 g $CO_{2\text{-eq}}$ MJ^{-1}, respectively.

When NTCFs are included (Fig. 2.d), the medians for FP, HTL, BSNG, and FT decrease to 54, 29, 3, and −6 g $CO_{2\text{-eq}}$ MJ^{-1}, mostly due to the inclusion of the cooling effects of NO_x released by biofuel combustion. As the characterization of climate impacts from NTCFs is inherently more uncertain than that of GHGs, the Monte Carlo analysis indicates larger uncertainty ranges, especially in the short-term (GWP20) where net climate impacts can be either positive or negative (Fig. 2b). The

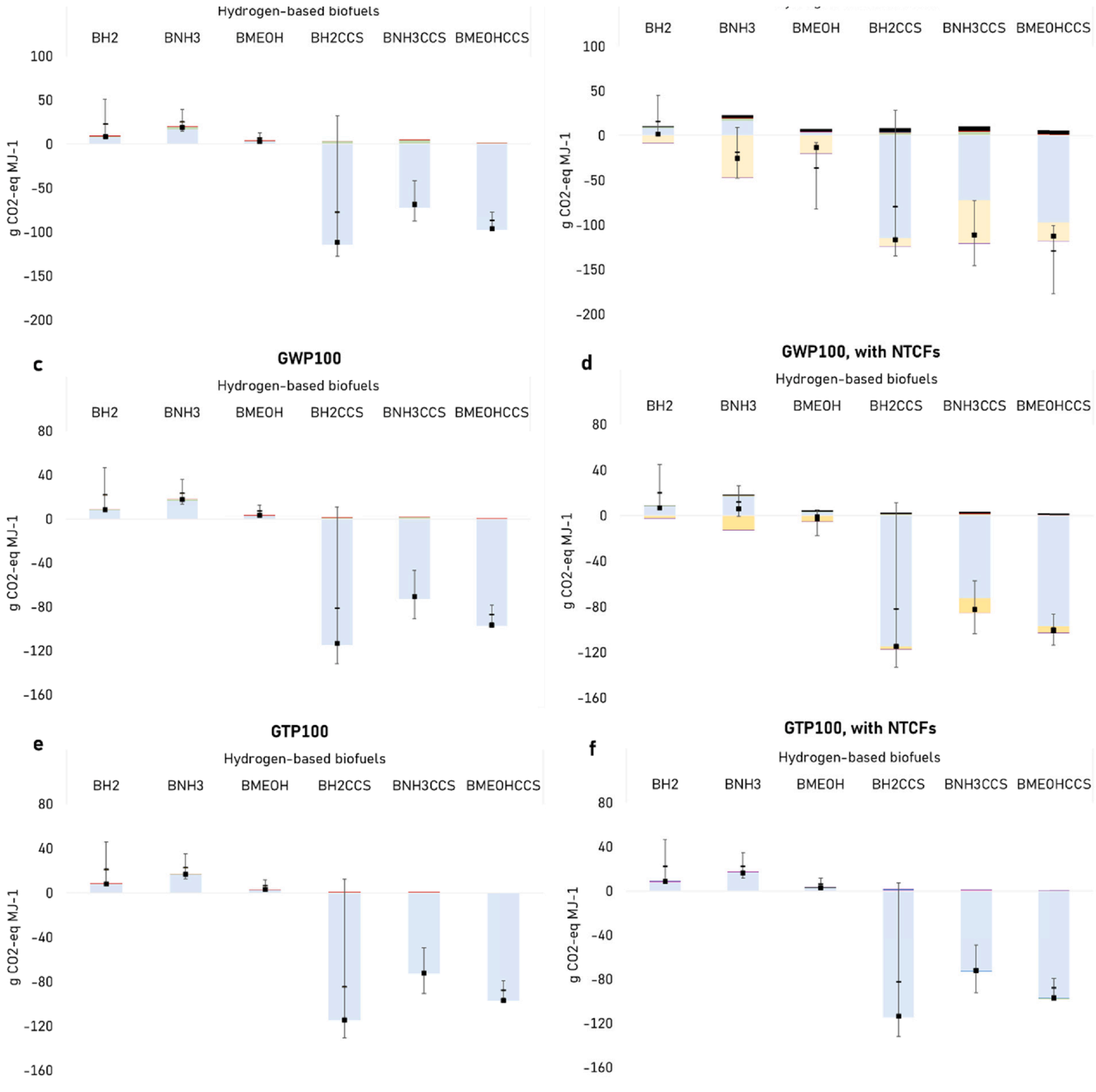


Fig. 3. Climate impacts of hydrogen-based biofuels from agricultural and forestry residues in Europe under multiple climate metrics with and without contributions of near-term climate forcers. Results are shown for GWP20 (a, b), GWP100 (c, d), and GTP100 (e, f). The biofuel pathways are the following: bio-hydrogen (BH2), bio-ammonia (BNH3), and bio-methanol (BMEOH) without and with carbon capture and storage systems (BH2CCS, BNH3CCS, BMEOHCCS). Breakdown and net impact (square-shaped point) refer to the deterministic analysis (det). Whiskers and lines show median, 5th, and 95th percentile confidence bounds from the Monte Carlo analysis, with variation in y-axis scales in the different panels.

lowest climate impacts are observed in the long term (GTP100) mainly because of the reduced importance of short-lived gases like methane, whereas CO_2 emissions dominate over other climate forcers for all biofuel pathways, as shown in Fig. 2e-f.

Among pathways associated with BECCS, negative GHG intensities are obtained even without considering the effects of NTCFs. FTCCS and BSNGCCS pathways have the lowest values – medians of −95 and −41 g $CO_{2\text{-eq}}$ MJ^{-1} with GWP100, respectively. As shown in Fig. 2c, such negative results are mainly due to the capture and storage of biogenic carbon dioxide from the biofuel plant. BSNGCCS presents a slightly higher impact because of the contribution of methane slips in the combustion stage and due to higher carbon venting from the power generation and gasification areas when compared to FTCCS. Even considering capture and storage, HTLCCS has a positive median of 22 $gCO_{2\text{-eq}}$ MJ^{-1} (Fig. 2c) because of two main reasons. First, the biofuel plant uses more electricity from the grid relative to other BECCS (mainly for the bio-crude upgrade), which contributes to increasing the net impacts when the average life cycle GHG emissions of the European grid – ca. 384 gCO_{2eq} kWh^{-1} – is considered. Second, a lower sequestration capacity (ca. 21 % of carbon in biomass) occurs in HTL than in the gasification-based routes, mainly because of the higher conversion of biomass into liquid biofuels; consequently, a lower share of the carbon input is transformed into gaseous byproducts (which contain carbon dioxide). HTLCCS has a wider range of uncertainties that can reach negative values for GWP100 only when biofuel plants are situated in European countries with more decarbonized electricity mixes. When NTCFs are included in the Monte Carlo analysis, the order of biofuels remains unchanged in terms of climate change impacts (Fig. 2b, d, and f), although slightly lower emissions and higher uncertainty ranges are observed due to the influence of NOx emissions in biofuel combustion.

3.2. Hydrogen-based biofuels

Fig. 3 shows the results of hydrogen-based biofuels. Bio-methanol (BMEOH) is the pathway with the lowest climate impact, with a GWP100-median and uncertainty range of 7 and 4–12 g $CO_{2\ eq}$ MJ^{-1}, respectively. This is mostly due to the lower electricity consumption from the grid than bio-ammonia (BNH3) and bio-hydrogen (BH2). Most of the impacts of BMEOH come from the transport of residues (ca. 50 %), with the remainder due to minor electricity imports from the grid and emissions from the distribution phase (see Fig. S1 in the Supplementary material). When comparing the results of BH2 and BNH3 (in GWP100), the Monte Carlo analysis indicates similar medians (22 and 23 g $CO_{2\ eq}$ MJ^{-1}, respectively). However, the net climate impact from the deterministic analysis considerably differs from the median in the BH2 scenario. This difference occurs due to assumptions made about hydrogen's downstream supply chain. As we assume the distribution of pressurized gaseous hydrogen in the deterministic scenario, lower electricity consumption is involved in the process of compression required for distribution. On the other hand, the uncertainty analysis includes the possibility of distributing liquid hydrogen, which pushes the median of climate impacts to a higher point. For this reason, BH_2 presents a wider range of impacts – from 9 to 47 g $CO_{2\ eq}$ MJ^{-1} – when compared to BNH3 (13 to 36 g $CO_{2\ eq}$ MJ^{-1}) also because of uncertainties associated with the different electricity mixes in Europe.

The influence of NTCFs on hydrogen-based fuels highlights the relevance of NO_x released in combustion as the main contributor to decreasing the net climate impacts (Fig. 3b, d). This effect is particularly relevant for methanol (BMEOH), whose median can reach negative climate impacts (-4 g $CO_{2\ eq}$ MJ^{-1}) even without considering CCS. In the case of the BH2 and BNH3, the inclusion of near-term climate forcers reduces the net impacts to 20 and 12 g $CO_{2\ eq}$ MJ^{-1}, respectively. It is important to highlight that the impacts presented in Fig. 3 consider emissions from internal combustion engines (ICE) for hydrogen [65,74] and ammonia [66], which are still under development and are likely to be commercially available by 2030 [4].

When hydrogen-based fuels are produced with CCS, negative GHG intensities result from carbon capture and storage in the biofuel plant. The pathway with the lowest deterministic climate impact is BH2CCS (-113 g $CO_{2\ eq}$ MJ^{-1}), followed by BMEOH and BNH3 (-96 and −70 g $CO_{2\ eq}$ MJ^{-1}, respectively) (Fig. 3c). Although all hydrogen-based CCS plants can store between 50 and 58 % of the biomass carbon input as carbon dioxide, the highest carbon capture capacity per energy output of biofuel is observed in BH2. This happens because this pathway has the lowest biomass-to-fuel energy efficiency, with approximately 36 % biomass-to-fuel conversion on an LHV basis (see Table S21 in the Supplementary material). When the uncertainties are included, higher electricity consumption associated with liquid hydrogen logistics increases the overall median of climate impacts of BH2 (to −81 g $CO_{2\ eq}$ MJ^{-1}) and favors the BMEOH pathway (median of −87 g $CO_{2\ eq}$ MJ^{-1}). For BNH3, both the deterministic and the median values are approximately the same (-70 g $CO_{2\ eq}$ MJ^{-1}). When NTCFs are included in the Monte Carlo analysis, the rank of hydrogen-based biofuels' remains unchanged (Fig. 3b, d, and f), although slightly lower climate impacts and higher uncertainty ranges are observed due to the inclusion of NOx.

3.3. FuelEU maritime goals and prospective LCA

3.3.1. Drop-in biofuels' impacts at a country level

Considering the large uncertainty of climate impacts at the European level, we performed a disaggregated analysis to identify the different potentials in each country to achieve the FuelEU Maritime reduction goals in terms of GHG intensity [2]. Fig. 4 shows the combinations of countries and biofuel options in Europe that can successfully meet these goals. Countries with colors (not gray) are those that reach the FuelEU Maritime target at the given year. The magnitude of the achieved GHG intensity reduction is indicated by the intensity of the color (the darker the color, the larger the climate change mitigation relative to fossil fuels). We also present how future improvements associated with REMIND SSP2 National Policies Implemented (NPi) scenario will influence the situation in each country up to 2050 when stricter mitigation potentials relative to fossil fuels are gradually required (7 % in 2030, 26 % in 2040, and 75 % in 2050).

For FP, 80 % of the EU countries would reach a minimum 7 % GHG intensity reduction by 2030, but countries with relatively large potentials (e.g., Germany, the Netherlands, Poland) will not meet this criterion unless climate policies are implemented (Fig. 4a). Over time, the dynamic targets from FuelEU Maritime limit FP pathways only to countries with cleaner electricity grids such as Norway, Sweden, France, and Switzerland by 2050 (Fig. 4a-c). These findings are strongly reliant on the bio-oil stabilization step, which requires hydrogen assumed to be produced from electrolysis using national electricity mixes. HTL (Fig. 4e-g) can reach reduction goals in all European countries by 2030, but a similar trend to FP is found when more strict targets are implemented.

The gasification-based biofuels (FT and BSNG) reach the GHG intensity reduction goals at any decade, even without any further technological improvements and efficiency gains derived from the implementation of the NPi scenario (Fig. 4d and 4 h). The average reductions in GHG intensity of BSNG and FT are in the range of 83–86 % and 94–98 %, respectively, since those biofuel plants do not require relevant external electricity from the grid. Since most of the impacts from gasification are related to biomass transport, the lowest emissions are observed in countries with higher average residue density per area such as Denmark, Germany, Hungary, the Czech Republic, and France. The opposite situation is observed in Norway, Portugal, Ireland, Sweden, Finland, Greece, and others where longer average transport distances are needed for transporting residues to the biofuel plants. However, although in a more limited number of countries, the overall GHG intensity reduction of HTL and FP can be higher than BSNG where the electricity mix is highly decarbonized – which is the case in Norway, Sweden, and France.

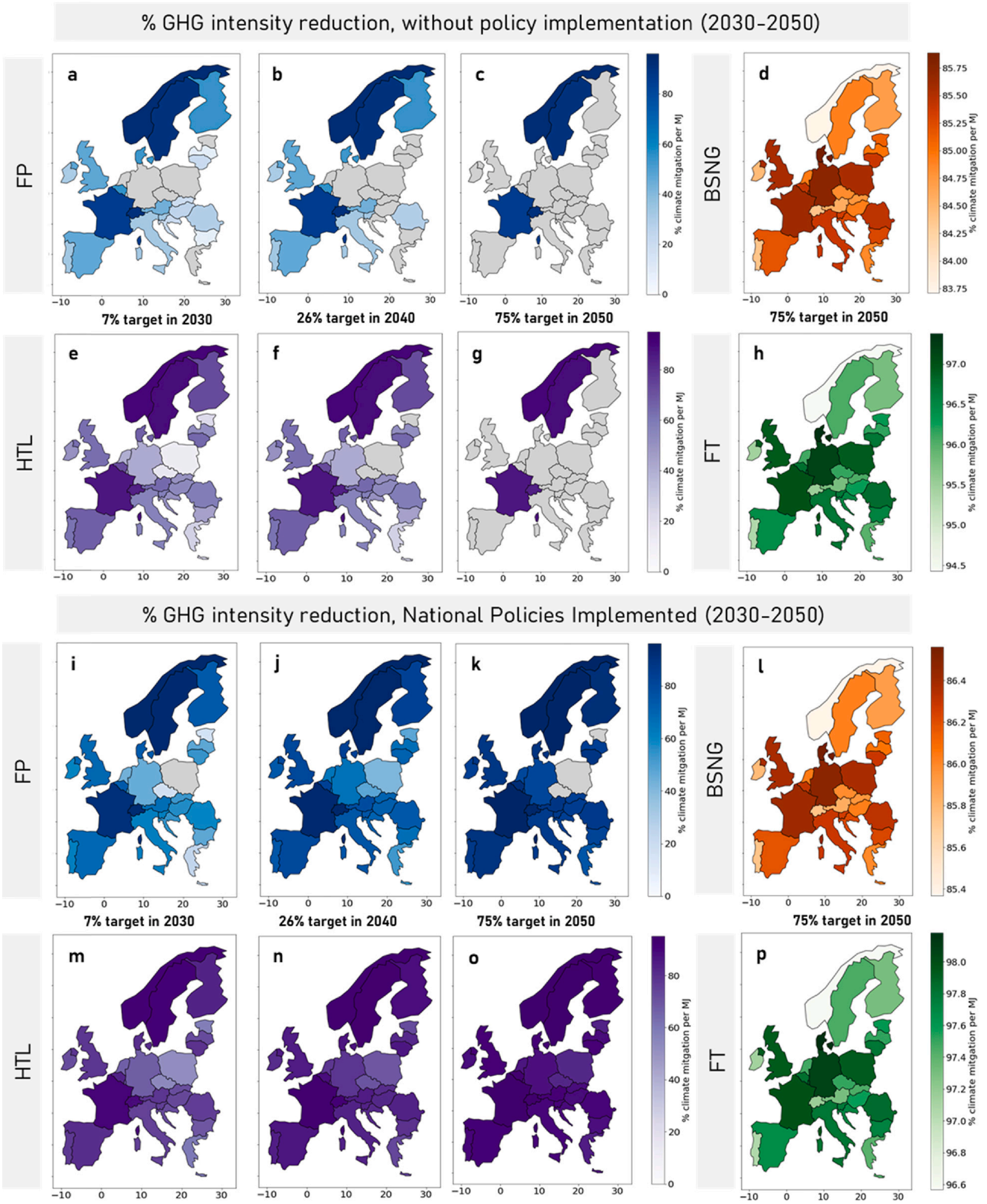


Fig. 4. Percentage reductions in GHG intensity of fast pyrolysis (a, b, c), bio-synthetic natural gas (d), hydrothermal liquefaction (e, f, g), and Fischer-Tropsch (h) marine biofuels from agricultural and forestry residues in different European countries relative to fossil fuels under dynamic FuelEU Maritime's goals (with or without climate policy implementation). The forecasted percentage reductions in FP (a-c), BSNG (d), HTL (e-g), and FT (h) are based on a constant technological and socio-economic background. Those for FP (i-k), BSNG (l), HTL (m-o), and FT (p) consider the changes in technological and socio-economic conditions that are consistent with the climate policies from REMIND SSP2 National Policies Implemented (NPi) until 2050. Countries highlighted in gray refer to those not achieving the minimum reductions according to Policy Option 3 [2] in comparison to the GHG intensity of the fossil fuel mix (87 g CO_{2eq} MJ^{-1}). Results refer to the deterministic analysis. Note the different scales in the climate change mitigation axes (the darker the color, the larger the climate change mitigation relative to fossil fuels).

The implementation of the climate pledges in the National Policies Implemented (NPi) under the SSP2 scenario brings technological and socio-economic improvements that affect the climate impact performances of marine biofuels (Fig. 4i-p). There are progressing reductions in GHG intensity of FP when shifting from the present to the future because of the continuous background improvements – especially the decarbonization of the electricity grids in European countries (see Table S35 in the Supplementary Material) – that are instrumental to drive biofuel's mitigation potential above the reduction targets in all countries in 2030 and most of them in 2050 (Fig. 4i-k). Countries excluded are Poland, the Czech Republic, and Estonia, which achieve mitigation of 70 %, 72 %, and 74 %, respectively – very close to the 75 % goal. This mitigation could increase in case of improvements in the biofuel conversion efficiencies, which are not considered in the deterministic analysis on which Fig. 4 is based. Evolutions in biomass-to-fuel conversion in the biofuel plant and enhanced efficiency in the electrolyzers over the next decades could lower the use of energy in the biofuel plant, thus making the projected GHG intensity reduction even more intense in all pathways. In the case of HTL (Fig. 4m-o), all future targets are achieved with the NPi scenario. In addition to the electricity, impact reductions associated with advances in the background activities, such as the production of chemicals – mainly potassium carbonate and sodium hydroxide used as inputs to the HTL process – significantly contribute to decreasing emissions as cleaner sources of heat and power generation are progressively implemented in the chemical industry. The gasification pathways (BSNG and FT) present lower sensitivity to the improvements of NPi scenario (Fig. 4l-p); therefore, there are only minor decreases in their GHG intensity that are mostly related to the evolution in transport systems that gradually benefit from increases in fleet fuel efficiency and progressing declines in fossil fuel use. In the case of BSNG, improvements are limited by methane slips during combustion which is assumed to remain unchanged in the future.

Drop-in biofuels associated with CCS systems are expected to deliver negative emissions, i.e., GHG intensity reductions above 100 % relative to fossil-based marine fuels. However, without the NPi scenario, HTLCCS is not able to fully deliver negative emissions in entire Europe and can only deliver the 75 % reduction goals by 2050 in about half of the countries (see Fig. S2 in the Supplementary Material). The full achievement of FuelEU Maritime reduction goals in Europe occurs only under the NPi scenario when the improvements in the supply chain will lead to cuts within the range of 108–122 % by 2050. The lowest emissions are associated with countries with decarbonized electricity mixes such as Norway, Sweden, and France, whose reductions are above 120 %. In the case of BNSG and FTCCS, both a higher share of carbon capture in the biofuel plant and lower use of external electricity in the biofuel plant contribute to negative emissions even without the further implementation of policies in Europe; the average GHG intensity reductions without a policy are in the range of 148–150 % and 213–216 %, respectively. Gasification-based pathways are less sensitive to the progressive improvements associated with NPi scenarios, with BNSGCCS and FTCCS reaching slight improvements to 151 % and 218 % by 2050, respectively; in both cases, the lowest average emissions are associated with countries with shorter biomass transport distances such as Denmark, Germany, Hungary, the Czech Republic, and France.

3.3.2. Hydrogen-based biofuels' impacts at a country level

Fig. 5 shows the average percentage reductions from hydrogen-based biofuels in each country under the deterministic approach. For pressurized bio-hydrogen (BH2-P), the average mitigation values without policy implementation range from 80 to 96 % and consider the distribution of compressed hydrogen, thus resulting in lower electricity use from the grid in comparison to its liquid form. BH2-P has its highest average reduction in France and Sweden (up to 96 % under current technology and up to 98 % under the NPi scenario), two countries that combine both more decarbonized electricity grids and higher density of residues per area (Fig. 5a, e). For liquid hydrogen (BH2-L), Fig. 5b shows that the additional electricity requirements make this pathway more distant from achieving the 75 % reduction goal by 2050 in most of Europe; on the other hand, when the NPi scenario is implemented (Fig. 5e), BH2-L reaches the goal established by the regulation in all European countries assessed in this study thanks to a cleaner future electricity system. In the case of BNH3 (Fig. 5.c), there is a similar trend, but relatively higher electricity consumption in the biofuel plant leads to insufficient GHG emission reduction by the 2050 s in countries like

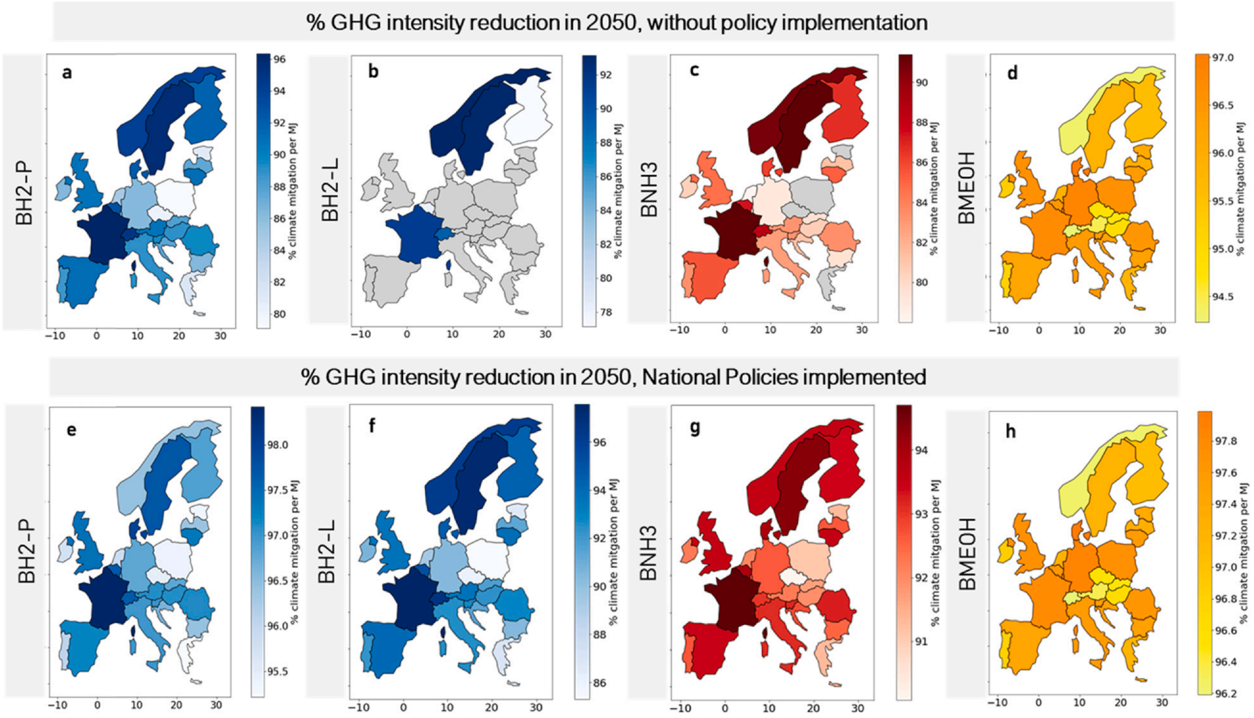


Fig. 5. Percentage reductions in GHG intensity of bio-hydrogen (a), bio-ammonia (b), and bio-methanol (c) from agricultural and forestry residues in different European countries relative to fossil fuels under the FuelEU Maritime's goals of a 75 % cut in GHG intensity by 2050 (without climate policy implementation). The forecasted percentage reductions for BH2 (d), BNH3 (e), and BMEOH (f) to current fossil fuel GHG intensity consider technological improvements from REMIND SSP2 National Policies Implemented (NPi) scenario in 2050. Countries highlighted in gray color refer to those not achieving the minimum reductions of 75 % in GHG intensity in comparison to the reference fossil fuel (87 g CO_{2eq} MJ^{-1}). Note the different scales in the climate change mitigation axes (the darker the color, the larger the climate change mitigation relative to fossil fuels).

Poland, the Czech Republic, Estonia, and Greece. This situation is reverted with the NPi scenario, with a European average percentage reduction of 90–95 % from bio-ammonia use in 2050 (Fig. 5g). Considering that BMEOH has the lowest electricity consumption and the highest biomass-to-fuel efficiency among the studied hydrogen-based pathways, Fig. 5d shows less variability in the scenarios without and with policy implementation: the average mitigation is 94–97 % and 96–98 %, respectively. In both cases, BMEOH has slightly lower emissions in Denmark, Germany, France, and Poland, which are the countries that combine both the highest average density of residues per area and shorter distances to the seaport, which minimizes the emissions associated with transport during the distribution phase.

In the case of BECCS, all hydrogen-based pathways can reach reductions above 100 % without further policy implementation, (see Fig. S3 in the Supplementary Material), even for the case of BNH3CCS and BH2CCS-L, whose lower values are 104 % and 116 %, respectively, when the Polish electricity grid is considered. The highest reduction potentials occurred under the BH2CCS-P scenario (ca. 258 %, in France, Sweden, and Switzerland under 2050′s NPi scenario); in the case of BMEOHCCS, the highest reductions in GHG intensity (217 %) are observed in France, Germany, Italy, Lithuania, Luxembourg, Romania, Slovenia and UK under 2050′s scenario with policy implementation.

3.3.3. *Mitigation and energy potentials at a European level*

An overview of GHG intensity potential reductions in Europe is shown in Fig. 6, where a broader comparison of both drop-in and hydrogen-based biofuels under a Monte Carlo analysis is provided for different mitigation thresholds. When considering the FuelEU Maritime initiative goals for the next decades, all technologies can likely meet the GHG intensity reductions by 2030, since all the medians are above the 7 % reduction benchmark even without policy implementation. Although drop-in biofuel pathways can technically reach values above 75 % by 2050, the highest uncertainties associated with pathways such as FP, HTL, and HTLCCS indicate that GHG intensities will be more dependent on the regional context (i.e., on the electricity mix of the country where they are produced). In the case of gasification technologies such as FT and BSNG, it is more likely that such pathways are less country-specific and stay between 75 % reduction and GHG intensity neutrality. Among drop-in biofuels associated with CCS, BSNGCCS and FTCCS are placed in the carbon-negative GHG intensity zone and are mostly dependent on the variability of the biomass-to-fuel conversion in the biofuel plant. HTLCCS median is also very close to the 2050 FuelEU target, however, such reductions are more consistent under the implementation of the NPi scenario by 2050. Among the drop-in alternatives, FP underperformed due to the assumptions made about hydrogen needs for bio-oil upgrading, but alternative sourcing – such as hydrogen from biogas – or even evolution in electrolyzer efficiencies, for instance, can importantly improve the results related to such biofuel pathway.

Bio-methanol has the lowest uncertainty range among hydrogen-based biofuels, and the median indicates a high probability of meeting well-to-wake GHG intensity reduction targets by 2050 even without further technological improvements. On the other hand, a wider range is observed for BH2 and BNH3 pathways with the influence of electricity consumption pushing their medians to levels below 75 % reduction by 2050; this indicates more dependency on the grid and on technological improvements over the next decades to deliver more prominent GHG intensity reductions. The widest uncertainty is associated with BH2CCS since it overlaps the risks associated with BH2 liquefaction, CCS electricity consumption, and the inefficiencies associated with liquid hydrogen distribution. Despite its large range, BH2CCS is more likely to deliver the highest maximum reduction among all biofuels in the carbon-negative GHG intensity zone if the NPi scenario is implemented. Without any further technological improvements, BMEOHCCS is associated with the lowest GHG intensity, even lower than BH2CCS. The pathway of bio-ammonia with carbon capture and storage also indicates consistency in the carbon-negative GHG intensity zone and its uncertainty range is slightly higher than BMEOH due to its superior electricity consumption per MJ of biofuel output.

Fig. 6 indicates a slight advantage towards the gasification-related pathways – mainly bio-synthetic natural gas, FT-diesel, bio-methanol, and all hydrogen-based biofuels produced with CCS –, since they are consistently above the 75 % GHG intensity reduction reference, even considering a lack of continuous implementation of environmental policies in European countries.

Fig. 7 offers a more thorough evaluation that combines local biofuel GHG intensity with the potential of fuel production per country depending on the availability of residues. For this reason, it provides an overview of the cumulative biofuel supply potential and greenhouse gas

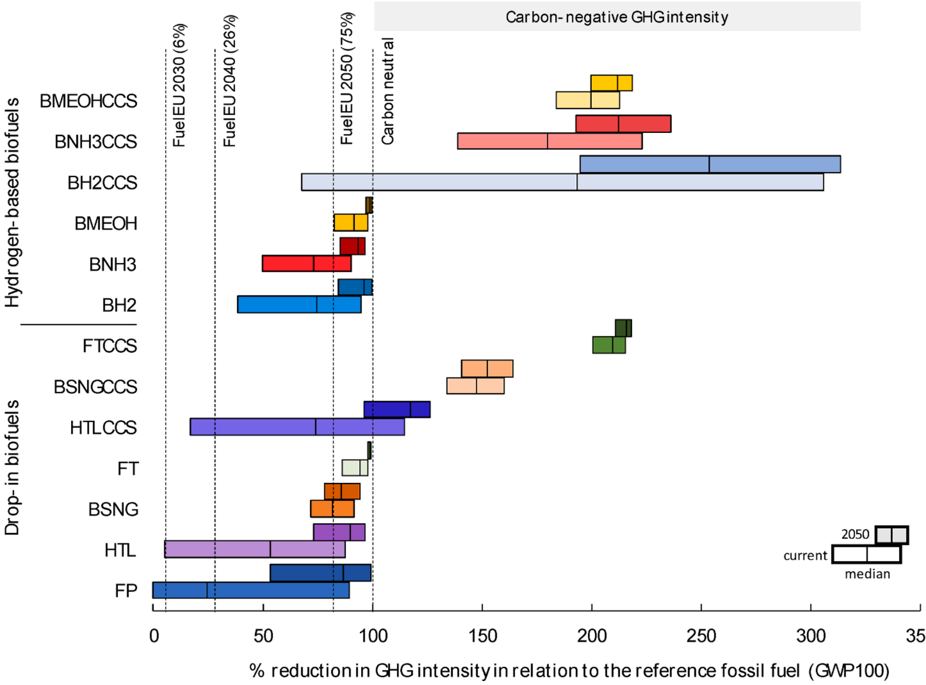


Fig. 6. Projected well-to-wake GHG intensity percentage reductions of biofuels from agricultural and forestry residues at an average European level in comparison to fossil fuel emissions [2] considering current and future (NPi 2050) scenarios. Results are for the following drop-in and hydrogen-based pathways: Fast Pyrolysis (FP), Hydrothermal Liquefaction (HTL), gasification to Bio-synthetic natural gas (BSNG), and Fischer-Tropsch (FT) without and with carbon capture and storage systems (HTLCCS, BSNGCCS, FTCCS); bio-hydrogen (BH2, both liquid and pressurized), bio-ammonia (BNH3) and bio-methanol (BMEOH) without and with carbon capture and storage systems (BH2CCS, BNH3CCS, BMEOHCCS). The box plots indicate the minimum, median, and maximum values from Monte Carlo analysis for current and future (NPi 2050) scenarios. The Monte Carlo analysis includes key uncertainties in the biofuel value chains, climate impacts, and the variability in the climate footprint of the electricity mixes in the different European countries.

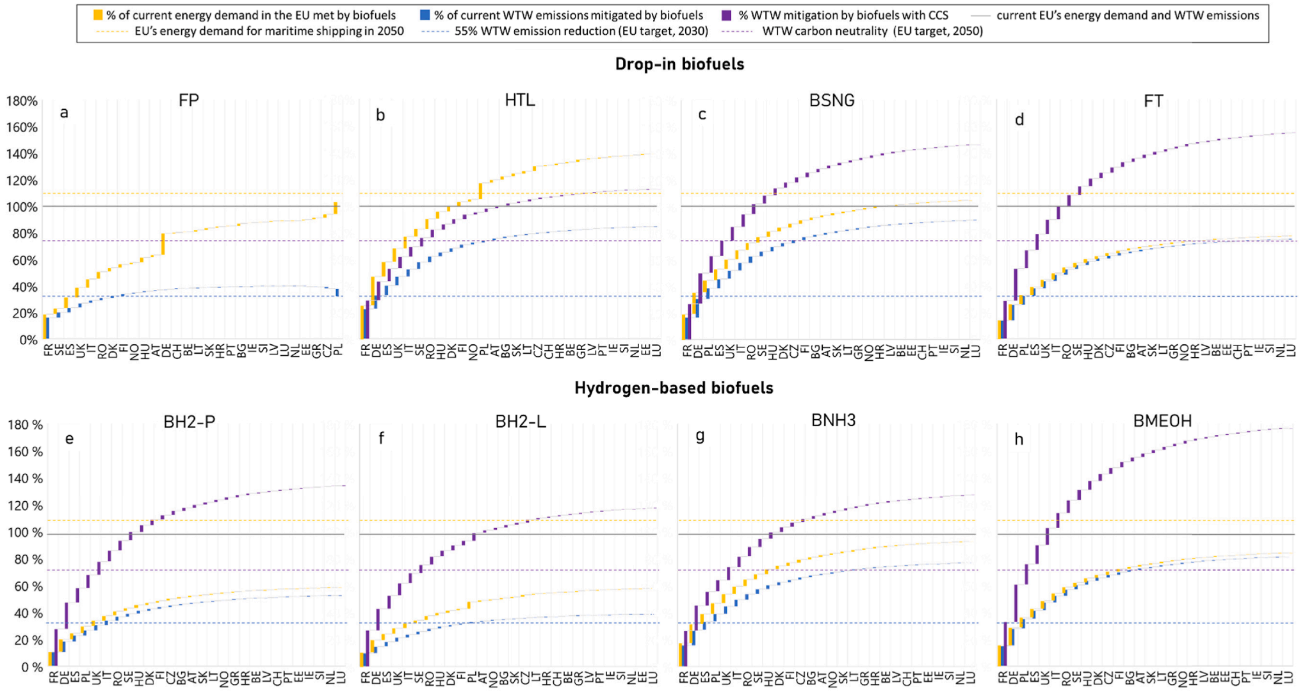


Fig. 7. Cumulative potentials of both GHG mitigation (blue and purple bars) and biofuel supply (yellow bars) per country using the current agricultural and forestry residues available in Europe. The maritime biofuels are FP (a), HTL (b), BNSG (c), FT (d), BH2-P (e), BH2-L (f), BNH3 (g), BMEOH (h), under current technology and background system (without NPi scenario). The gray line represents both the current fuel demand for EU maritime transport and the current well-to-wake (WTW) GHG emissions [2,41]. The yellow dashed line depicts the projected energy consumption for 2050 according to FuelEU Maritime Policy Option 3 [7]. The blue- and purple-dashed lines indicate the targets of 55% reduction and carbon neutrality for the European maritime sector to 1990 emissions, respectively [3].

mitigation per year when considering the contributions from the 30 European countries. The yellow bars in Fig. 7 show the bioenergy production potentials in each country if all agricultural and forestry residues currently available in Europe – under a sustainable removal scenario – were converted into biofuels using the current technology without any improvements in the supply chain structure. France, Germany, Italy, Poland, Romania, Spain, Sweden, and the UK stand out since they provide larger amounts of residues, therefore maximizing biofuel production when pathways with higher biomass-to-fuel conversion rates are considered. This is the case of FP, HTL, and BSNG (Fig. 7a-c), which can supply more than the current yearly energy demand in the EU Maritime transport sector [40]. HTL has the largest energy production potential among all drop-in and hydrogen-based pathways, providing up to 140 % of the current demand (Fig. 7b). France, Germany, and Poland together have the potential of delivering 60 % of the energy currently used in the EU's maritime sector. FT is the drop-in pathway with the lowest biomass-to-fuel conversion efficiency, thus providing three-quarters of the current maritime fuel demand (Fig. 7d). Likewise, the supply from hydrogen-based biofuel pathways does not fully achieve the current consumption in the EU, although biomethanol and bio-ammonia reach 84 % and 93 % of the European maritime energy demand, respectively (Fig. 7e-h).

Another possible benchmark to evaluate the potential of biofuels' energy supply is the projection from FuelEU Maritime's Policy Option 3 (PO3) which describes a gradual uptake of renewable and low carbon fuels (RLFs) in the total EU maritime fuel mix of up to 89 % by 2050 [40]. The intended values of drop-in and hydrogen-based biofuels would reach together a maximum of 65.6 % in the EU mix: the breakdown for biofuels, bio-LNG, hydrogen, ammonia, and methanol is 42.4 %, 15.4 %, 7.2 %, 0.4 %, and 0.2 %, respectively, in 2050. Under these assumptions, even a mix of biofuel pathways with the lowest biomass-to-fuel conversion efficiencies – such as FT and BH2-L – would supply the necessary energy demand by 2050 under PO3 when using the current agricultural and forestry residues available in Europe (see Table S36 in the Supplementary Material).

The cumulative GHG mitigation at the European level is indicated by the blue bars (Fig. 7), which combine country-specific biofuel production potentials – based on their specific availability of crop and agricultural residues – with their respective life cycle GHG intensities. GHG mitigation values are calculated in tons of $CO_{2\text{-eq}}$ per year and then normalized according to the total current well-to-wake emissions from the European maritime sector. As explained in the Methods (Section 2.3), we recalculate the EU maritime transport's emissions by including those from fuel production (well-to-tank). The result – 159 Mt CO_{2eq} yr^{-1} – considers the well-to-wake emissions based on the current fuel consumption and average GHG intensity of fossil fuel mix from EU Regulation 2017/352 [2]. Similarly, the European international shipping well-to-wake emissions in 1990 of 115 million tons CO_{2eq} per year refers to 103 Mt CO_{2eq} yr^{-1} from combustion [3,42] and 11.8 Mt CO_{2eq} yr^{-1} from fossil fuel production [2,41] (see parameters and calculations in Tables S3-S6 of the Supplementary Material). Therefore, the dashed-blue and dashed-purple lines represent a reduction of 55 % GHG emissions by 2030 and climate neutrality by 2050 in relation to 1990 emission levels [6], respectively. The full deployment of biofuels using agricultural and forestry residues in Europe can be an effective strategy to mitigate the overall emissions by the next decade, since all drop-in and hydrogen-based pathways achieve at least 55 % reduction by 2030, including FP (Fig. 7a) whose cumulative mitigation potential is particularly low due to little (or even negative) contributions from countries with more carbonized electricity grids. When it comes to the carbon neutrality goal set for 2050 in the EU maritime sector, HTL, BSNG, FT, BMEOH, and BNH3 have the highest potential of delivering such mitigation targets without using CCS technologies; among them, HTL and BNSG stand out (Fig. 7b and 7c).

The purple bars present the cumulative mitigation potentials when CCS is integrated into the biofuel plants. A situation with absolute carbon neutrality – i.e., zero net carbon emissions compared to the current situation – and beyond (e.g., carbon negative) can be achieved when coupling HTL, BSNG, and FT to CCS systems among drop-in biofuels. As highlighted in Fig. 7d, the maximum overall mitigation of 155 % is obtained from the FTCCS pathway among drop-in biofuels, even though the full deployment of such technology in Europe would mean the partial supply (78 %) of the current EU maritime fuel demand. The highest relative benefits from CCS implementation are observed in bio-hydrogen pathways (Fig. 7f-g), where the total mitigation potential can achieve a 3-fold increase in the case of BH2CCS-L (Fig. 7f). BNH3CCS (Fig. 7g) is also an interesting pathway due to its potential of combining both high energy supply (93 % of the current energy demand) and overall GHG mitigation (127 % of the current EU's maritime transport WTW emissions). On the other hand, if the objective is delivering the maximum overall decarbonization in the EU maritime sector, then all gasification-based pathways with CCS are promising options, highlighting bio-methanol (BMEOHCCS, Fig. 7h) with a potential of achieving 176 % mitigation.

3.3.4. *Synthesis*

It is essential to highlight the differentiation made between the results of GHG intensity (Figs. 4-6) and potential mitigation per year at a European scale (Fig. 7). The first approach measures the climate impact in g $CO_{2\text{-eq}}$ per MJ, whose values depend on both the biofuel pathway and country. This means that the best biofuel option – i.e., that presenting the lowest GHG intensity in a country – will necessarily result in the largest mitigation measured per year for that country. On the other hand, the potential mitigation per year (t CO_{2eq} yr^{-1}) at a European scale depends on both the GHG intensities and biomass residues availabilities; both are changing from country to country. Therefore, the second approach leads to a less obvious outcome when defining the best biofuel option, since the country with the lowest GHG intensity for a given biofuel does not necessarily have the highest biomass availability and industrial conversion yield to potentialize these mitigation effects per year.

Based on the results presented in Figs. 4-6, bio-synthetic natural gas, Fischer-Tropsch diesel, and bio-methanol provide the largest GHG intensity reductions (i.e., have the lowest g CO_{2eq} MJ^{-1}) with the current technology, thus achieving the FuelEU Maritime goals for the next decades even under less optimistic deployment of climate policies. On the other hand, a broader perspective toward full deployment of drop-in biofuels at a European scale with the current technology (Fig. 7) mainly favors bio-synthetic natural gas and hydrothermal liquefaction, since they provide the largest overall mitigation potentials (i.e., in tons $CO_{2\text{-eq}}$ per year) which represent 90 % and 84 % reduction of current European maritime transport emissions, respectively; Fischer-Tropsch diesel, bio-methanol, and bio-ammonia are also important alternatives since they meet the carbon neutrality target by 2050 in Europe without using CCS.

The climate mitigation benefits from the adoption of bio-hydrogen, particularly in the liquid form, will still depend on further advances in the entire production chain mainly to decrease the carbon intensity of the electricity grid and on improvements in the efficiencies associated with storage, distribution, and bunkering of hydrogen on a larger scale. Although our results for carbon-free RLFs favored bio-ammonia as a more efficient carrier of hydrogen at the European scale under the current technology status, the uncertainty analysis for all biofuels indicates that the largest individual GHG intensity reductions are obtained with bio-hydrogen, especially when considering the projected decarbonization of the electricity grids until 2050.

The full deployment of carbon capture and storage proved to represent an opportunity to accelerate the pace of decarbonization in the European maritime sector since they hold the capacity of almost tripling the mitigation potential of biofuels without CCS, with GHG intensity reduction especially amplified when connected with bio-hydrogen production. At the European scale, however, the full deployment of bio-methanol with CCS achieved the largest GHG mitigation potential among all advanced biofuel pathways.

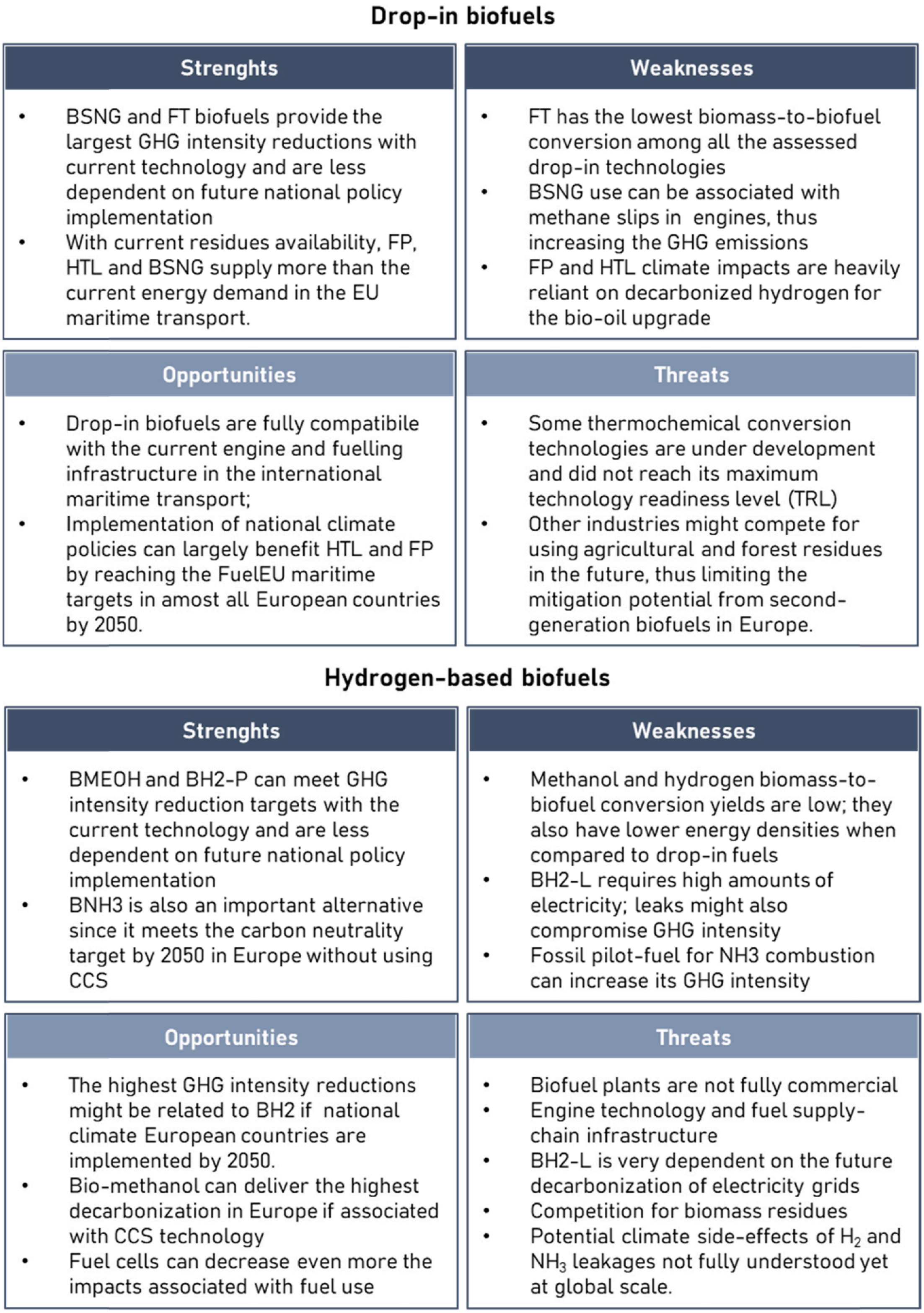
The main advantages and disadvantages of both drop-in and hydrogen-based biofuels based on the SWOT (Strengths, Weaknesses, Opportunities, and Threats) analysis are shown in Fig. 8, where the main implications of the different choices are reviewed in terms of their potential climate impacts. Techno-economic, safety, standard, and other important aspects associated with the implementation of such biofuel options at a larger scale are not included in this analysis, and we refer to other studies [11–13].

The main strengths and opportunities have been discussed with the results above, with an identification of the weaknesses that limit mitigation benefits for each technological option. Regarding the threats to the climate mitigation potentials of biofuels, they are largely connected to the technology readiness level (TRL) of current thermochemical options, which are still in the range of TRL 6–8 [75], not achieving the full commercial scale yet (TRL 9). This could negatively impact the deployment of the expected energy and climate mitigation from biofuels at the European scale in the short-term. Another relevant aspect is the current use of both crop and forest residues by other industries in Europe [38,39] or by increasing competition for biomass resources as other sectors need to find alternative renewable options to fossil fuels (e.g., aviation, cement, metallurgical industries, etc.). This might lead to increases in prices of biomass raw materials and limit large-scale deployment of biofuel plants and their associated climate mitigation benefits, as biomass and land resources are limited, and their sustainable supply needs to be regulated. In the case of hydrogen-based fuels, threats are associated with possible hydrogen leakages into the atmosphere. Considering that the hydrogen supply chain is still under development, future leakage rates associated with the production, storage, distribution, and use are still unknown and might also indirectly affect the climate [76]. The same applies to ammonia, since a large-scale ammonia-powered shipping trend could induce a fourfold increase of reactive nitrogen into the environment [77], with unknown consequences on the climate system at global scale. Moreover, the emissions related to use of ammonia in internal combustion engines can be affected by the type of pilot fuel used to improve ignition properties, which can represent about 5 % of total heat input energy [78].

4. Conclusions

Both drop-in and hydrogen-based biofuels will be essential for decarbonizing the EU maritime transport sector. Many successful combinations of biofuel options and countries have the potential to achieve the FuelEU Maritime targets today and in 2050. Drop-in biofuels have many advantages in the current technological scenario given the compatibility with the available engine technology and fueling infrastructure in international maritime transport. Although the climate impacts of some drop-in biofuels (e.g., those from HTL and FP) are very sensitive to the hydrogen supply chain and electricity mix of each European country, they have the largest potential to meet the fuel energy demand from the EU maritime transport (up to 140 %). On the other hand, hydrogen-based biofuels have a lower energy potential but in the long-term they can improve the climate performance of biofuels as they achieve the lowest GHG intensity per unit of fuel. Their capacity for decarbonization can be largely amplified with a more mature supply chain structure, which will certainly benefit from the implementation of national policies aiming at using more renewable energy sources over the next years.

With the current technology, BNSG, FT, and BMEOH pathways offer the largest GHG intensity reductions per unit of energy, and the full deployment of biofuels at the European scale favors BNSG and HTL because of their higher overall GHG mitigation. Among carbon-free biofuels, BNH3 can lead to higher mitigation currently, but the largest individual GHG intensity reductions in the future are achieved with BH2, especially with the projected decarbonization of the electricity grids until 2050. The full deployment of CCS represents an opportunity to accelerate the pace of decarbonization in the maritime sector,



Drop-in biofuels

Strenghts

- BSNG and FT biofuels provide the largest GHG intensity reductions with current technology and are less dependent on future national policy implementation
- With current residues availability, FP, HTL and BSNG supply more than the current energy demand in the EU maritime transport.

Weaknesses

- FT has the lowest biomass-to-biofuel conversion among all the assessed drop-in technologies
- BSNG use can be associated with methane slips in engines, thus increasing the GHG emissions
- FP and HTL climate impacts are heavily reliant on decarbonized hydrogen for the bio-oil upgrade

Opportunities

- Drop-in biofuels are fully compatibile with the current engine and fuelling infrastructure in the international maritime transport;
- Implementation of national climate policies can largely benefit HTL and FP by reaching the FuelEU maritime targets in amost all European countries by 2050.

Threats

- Some thermochemical conversion technologies are under development and did not reach its maximum technology readiness level (TRL)
- Other industries might compete for using agricultural and forest residues in the future, thus limiting the mitigation potential from second-generation biofuels in Europe.

Hydrogen-based biofuels

Strenghts

- BMEOH and BH2-P can meet GHG intensity reduction targets with the current technology and are less dependent on future national policy implementation
- BNH3 is also an important alternative since it meets the carbon neutrality target by 2050 in Europe without using CCS

Weaknesses

- Methanol and hydrogen biomass-to-biofuel conversion yields are low; they also have lower energy densities when compared to drop-in fuels
- BH2-L requires high amounts of electricity; leaks might also compromise GHG intensity
- Fossil pilot-fuel for NH3 combustion can increase its GHG intensity

Opportunities

- The highest GHG intensity reductions might be related to BH2 if national climate European countries are implemented by 2050.
- Bio-methanol can deliver the highest decarbonization in Europe if associated with CCS technology
- Fuel cells can decrease even more the impacts associated with fuel use

Threats

- Biofuel plants are not fully commercial
- Engine technology and fuel supply-chain infrastructure
- BH2-L is very dependent on the future decarbonization of electricity grids
- Competition for biomass residues
- Potential climate side-effects of H_2 and NH_3 leakages not fully understood yet at global scale.

Fig. 8. SWOT analysis applied to both drop-in and hydrogen-based biofuels from crop and forest residues in the European context.

especially when associated with bio-methanol. Depending on technical, economic, and other eventual constraints, a combination of different biofuel pathways tailored to the characteristics of the different countries (in terms of resource availability and distribution and electricity mix) can be strategically planned at a European level to optimize the climate change mitigation potential that can be achieved in the maritime transport sector.

The present study has limitations that can be addressed in future works. For example, estimates of biomass residue availability potentials in Europe can change in the future, and their projections can be harmonized with each specific SSP using IAMs. The analysis can also explore how the energy potentials are affected by competition for biomass residues from other sectors such as heat and electricity generation, aviation, and others. However, these aspects will affect the

European potential but not the specific climate change benefits of a given technology quantified in this study, which are rather sensitive to the possible evolutions in conversion technologies and efficiencies.

Choosing the most suitable RLFs from a regional perspective is perhaps just the start of a complex transition roadmap towards carbon neutrality in international maritime transport. Safety aspects and other challenges associated with a transition to a new supply chain infrastructure will be also important, as well as a coordinated action among countries fusing sustainable use of natural and technological resources with a consistent implementation of national policies. These factors combined will be crucial to match the large-scale uptake of RLFs with the expectations made on their decarbonization potentials for the next decades at a European level.

CRediT authorship contribution statement

Marcos D.B. Watanabe: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Writing – original draft. **Francesco Cherubini:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision. **Alexandre Tisserant:** Software, Validation, Writing – review & editing. **Otavio Cavalett:** Conceptualization, Methodology, Investigation, Resources, Writing – review & editing, Supervision.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

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