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Environmental trade-offs of renewable jet fuels in Brazil: Beyond the carbon footprint

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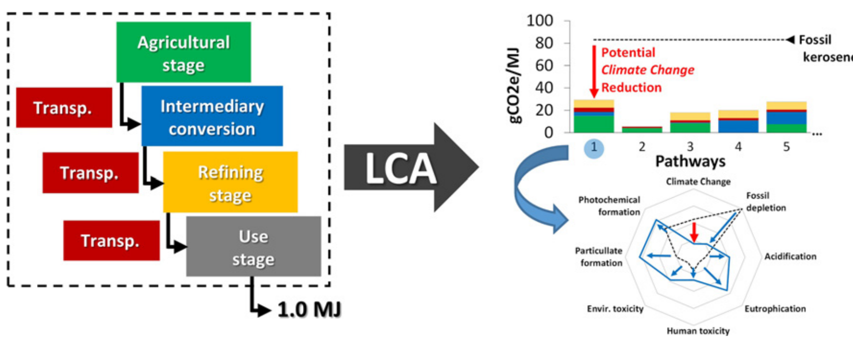
HIGHLIGHTS

- Life cycle assessment of renewable jet fuels produced by 1G/2G pathways in Brazil
- Climate change and seven other impacts categories were analyzed.
- 1G pathways can lead to 65% GHG emissions reduction, while 2G's, to more than 67%.
- Use of residues as feedstock does not necessarily lead to better environmental performance.
- Trade-offs among the impact categories are highly influenced by the upstream stage.

GRAPHICAL ABSTRACT



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ABSTRACT

The use of renewable jet fuels (RJFs) is an option for meeting the greenhouse gases (GHG) reduction targets of the aviation sector. Therefore, most of the studies have focused on climate change indicators, but other environmental impacts have been disregarded. In this paper, an attributional life cycle assessment is performed for ten RJF pathways in Brazil, considering the environmental trade-offs between climate change and seven other categories, *i.e.*, fossil depletion, terrestrial acidification, eutrophication, human and environmental toxicity, and air quality-related categories, such as particulate matter and photochemical oxidant formation. The scope includes sugarcane and soybean for first-generation (1G) pathways and residual materials (wood and sugarcane residues, beef tallow, and used cooking oil-UCO) for second-generation (2G) pathways. Three certified technologies to produce RJF are considered: hydroprocessed esters and fatty acids (HEFA), alcohol-to-jet (ATJ), and Fischer-Tropsch (FT). Assuming the residual feedstocks as wastes or by-products, the 2G pathways are evaluated by two different approaches, in which the biomass sourcing processes are either accounted for or not. Results show that 1G pathways lead to significant GHG reductions compared to fossil kerosene from 55% (soybean/HEFA) to 65% (sugarcane/ATJ). However, the sugarcane-based pathway generated three-fold higher values than fossil kerosene for terrestrial acidification and air quality impacts, and seven-fold for eutrophication. In turn, soybean/HEFA caused five-fold higher levels of human toxicity. For 2G pathways, when the residual

Abbreviations: 1G, first-generation; 2G, second-generation; 2Gh, second-generation ethanol from enzymatic hydrolysis; 2Gs, second-generation ethanol from syngas fermentation; ATJ, Alcohol-to-Jet; CHP, Combined Heat and Power; DCSH, direct conversion of sugar to hydrocarbons; FT, Fisher-Tropsch; GHG, greenhouse gases; HEFA, hydroprocessed esters and fatty acids; LCM, lignocellulosic material; LUC, land use change; RJF, Renewable Jet Fuel; SC, sugarcane; SMR, steam methane reform; UCO, used cooking oil; VSB, Virtual Sugarcane Biorefinery; WE, water electrolysis; WO, wood residues.

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feedstock is assumed to be waste, the potential GHG emission reduction is over 74% with no relevant trade-offs. On the other hand, if the residual feedstocks are assumed as valuable by-products, tallow/HEFA becomes the worst option and pathways from sugarcane residues, even providing a GHG reduction of 67% to 94%, are related to higher impacts than soybean/HEFA for terrestrial acidification and air quality. FT pathways represent the lowest impacts for all categories within both approaches, followed by UCO/HEFA.

1. Introduction

The international civil aviation sector has set ambitious targets to achieve carbon-neutral growth from 2020 and reduce its greenhouse gas (GHG) emissions by 50% by 2050 relative to 2005 levels. (ICAO, 2016). The renewable jet fuels (RJFs) is an important means of achieving these targets (ICAO, 2017), being used as drop-in fuels blended with fossil kerosene. The technologies used to produce RJFs fall into three groups (Cortez et al., 2014): lipid conversion (Pearlson, 2011), thermochemical, (Klerk, 2011) and biochemical processes (Moreira et al., 2014; Staples et al., 2014). From these three groups, five technologies have been approved by the ASTM (2019) with different blending restrictions: hydrotreating oil-based feedstocks (hydroprocessed esters and fatty acids, HEFA), dehydration and oligomerization of iso-butanol or ethanol (alcohol-to-jet, ATJ), direct conversion of sugar to hydrocarbons (DCSH), and the Fischer-Tropsch (FT) process.

According to Dodd (2018), more than 140 thousand commercial flights have been supplied by RJF since 2011. It corresponds to a sharp increase of RJF production, which achieved 13 million liters in 2018, and accounts for 6 billion liters in future purchased agreements. However, an accelerated deployment of sustainable biofuels is required to reach low carbon scenarios in the coming decades (Feuvre, 2018), with competitive costs and meeting sustainability standards. In this context, Brazil is considered as a potential supplier of RJF because of its large biomass production and technical experience in bioenergy (Cortez et al., 2014). Currently, sugarcane ethanol represents almost 20% of the country's road transport fuel consumption, while biodiesel, mostly from soybean oil, accounts for 10% of diesel consumption (EPE, 2019). At the same time, the use of residues, such as crop residues and waste greases, as energy source is already in place in Brazil. These promising feedstocks are well accepted as GHG mitigation strategy due to no relevant concerns related to land use change (LUCs) and food competition aspects (European Parliament, 2009; U.S. EPA, 2010a). For example, sugarcane bagasse supplies around 6% of Brazil's electricity demand (*i.e.*, 35 GWh) and waste greases, such as used cooking oil (UCO) and beef tallow, represents 18% of Brazilian biodiesel production (EPE, 2019). Furthermore, the 33.5 million tons of wood residue available in 7.7 million ha of planted forests (IBA, 2017), along with bagasse surplus and sugarcane cane straw, are potentially relevant feedstocks for bioenergy production in Brazil, including RJF.

With respect to the environmental performance of products, the life cycle assessment (LCA) has been a frequently employed tool for the evaluation of different environmental impact categories (Corbella et al., 2017; Zhang et al., 2019). Specifically for the aviation industry, the GHG reduction potential of several RJF pathways has been widely reported in the literature (Cavalett and Cherubini, 2018; de Jong et al., 2017; Han et al., 2013, 2017; Klein et al., 2018; Seber et al., 2014; Staples et al., 2014) due to the current sectorial goals However, the environmental effects and the possible trade-offs between different environmental impact categories along the RJF life cycle remain rather unexplored. Staples et al. (2013) evaluated the water footprint of middle distillate fuels in the United States. In Australia, Cox et al. (2014) reported the environmental performance of RJF from microalgae, Pongamia oil, and sugarcane molasses by eutrophication, water, land, and fossil energy use. In turn, Li and Mupondwa (2014) evaluated the jet fuel and biodiesel from camelina oil in Canadian Praires under endpoint impact categories, such as global warming potential, human health, ecosystem quality, and energy resource consumption. On the other hand, Klein et al. (2018) discussed the benefits of different routes for producing RJF by integrated designs to sugarcane mills in Brazil, considering environmental aspects related to human toxicity, terrestrial acidification, agricultural land occupation, fossil depletion, and climate change. Finally, Cavalett and Cherubini (2018) analyzed RJF production from forest residues in Norway for climate change mitigation and other environmental issues, which are embraced within the context of the Sustainable Development Goals (SDGs) (UN, 2017).

Even so, these analyses are scope-limited by either considering few categories or a small number of technical options, making it difficult to assess the environmental trade-offs of RJF production in different technical contexts.

In this sense, this paper aims to contribute to this research gap carrying out a harmonized and detailed LCA of ten strategic RJF pathways in Brazilian conditions and pointing out the possible trade-offs between the different impact categories. The pathways – which were represented by literature, modeling, and first hand-data, and local-specific life cycle inventories – comprised three ASTM-approved jet fuel-technologies (HEFA, ATJ, and FT) and six different feedstocks. The production systems were categorized as first-generation (1G) pathways – *i.e.*, from food-based feedstocks, such as soybean oil and sugarcane – and second-generation (2G) pathways, *i.e.*, from residue-based feedstocks, such as beef tallow, UCO, sugarcane residues and forestry residues, which were compared with each other and with fossil kerosene (Jet A).

2. Methods

The LCA was carried out considering the following steps, as recommended by the ISO (2006).

2.1. Goal and scope definition

A *well-to-wake* analysis – *i.e.*, from feedstock production to RJF use in aircraft – was performed by attributional approach, which focuses on the environmentally-relevant physical flows described by averaged data to and from the product-system (JCR, 2010). The functional unit was 1.0 MJ_{RJF} of energy supplied to aircraft.

2.1.1. System boundaries

The product-system for each RJF pathway was depicted in four stages, as presented in Fig. 1 and detailed in Section 2.2. The “upstream stage” is related to the feedstock sourcing and its treatment (*e.g.*, agricultural processes, feedstock collection, cattle management, and slaughterhouse). The “midstream stage” refers to feedstock processing into intermediary products for RJF conversion, which takes place at the “downstream stage”. Finally, the “use stage” involves RJF combustion in aircraft engines. The transportation between each stage is also considered. Jet A is used as the benchmark for comparative purposes.

Notwithstanding the environmentally sound appeal of using waste as a feedstock, it is frequently argued that whether such materials should still be regarded as waste as their utilization gains relevance, while, in some instances, alternative uses may already be in place. In the face of the rather arbitrary definitions around waste and by-

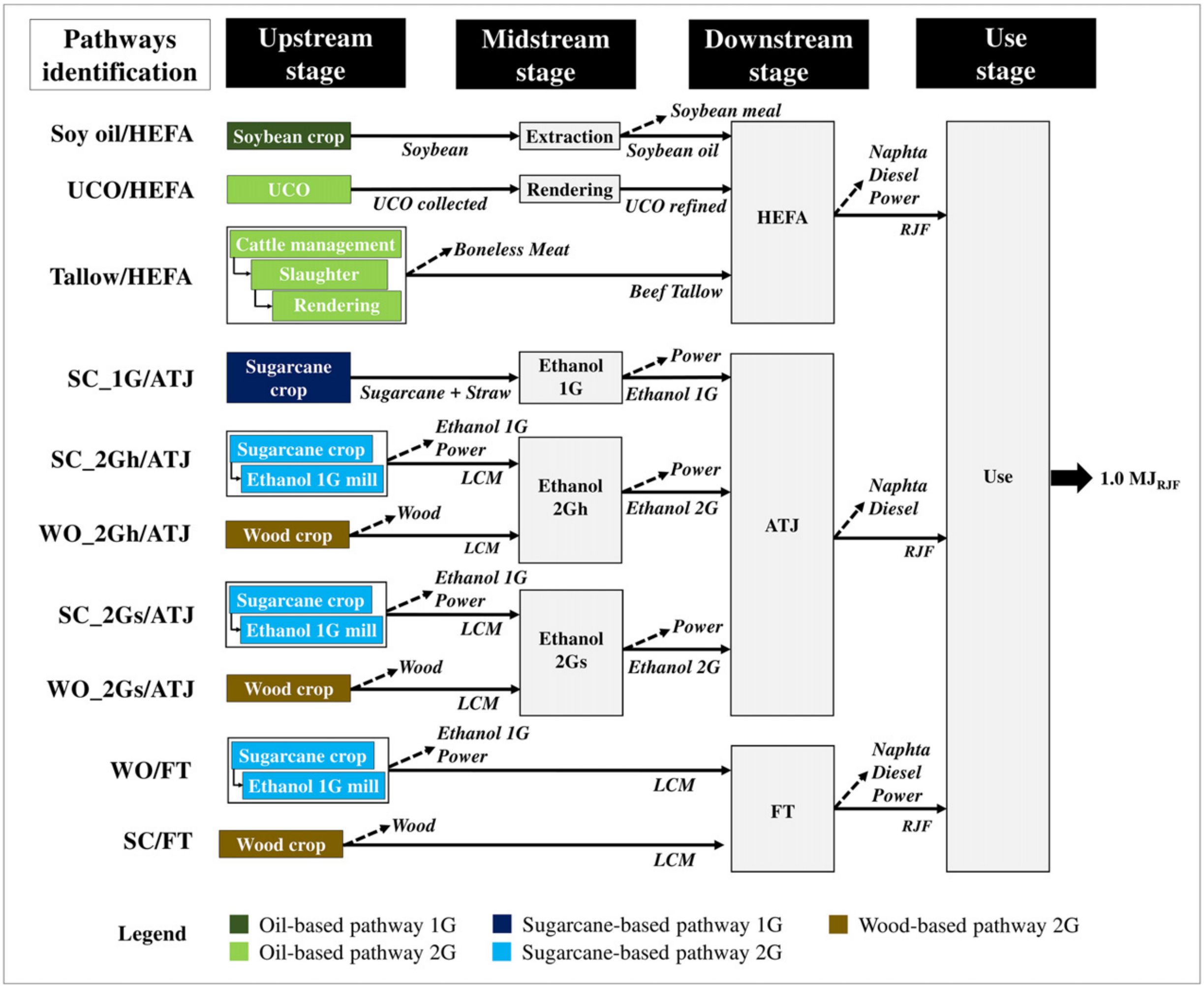

Fig. 1. Life Cycle stages for RJF production. Feedstocks: UCO, used cooking oil; SC, sugarcane; LCM: lignocellulosic material. Midstream stage: 1G, first-generation ethanol mill; 2Gh: second-generation ethanol mill from enzymatic hydrolysis; and 2Gs: second-generation ethanol mill from syngas fermentation. Downstream stage: HEFA, Hydroprocessed esters and fatty acids; ATJ, Alcohol-to-Jet; and FT: Fisher-Tropsch. Dotted lines for by-products.

products, two different approaches were considered for the residue-based pathways.

In System 1 (S1), residual feedstocks are deemed as waste, hence sugarcane and wood residues, beef tallow, and UCO do not carry a burden related to their generation. This approach has already been applied in low-carbon policies – such as the Renewable Energy Directive (European Parliament, 2009) in Europe and the Renovabio in Brazil (ANP, 2018). The methodology of the Renewable Fuel Standard program (U.S. EPA, 2010b) in the United States accounts for only the environmental burdens of the upstream stages related to nutrient compensation due to the crop residues' removal from the field and those related to the rendering process for tallow. Here, nutrient compensation was considered as a consequence of a decision, then it is not accounted for within a strict attributional approach. For tallow-based pathways, the rendering plant was assumed to be attached and integrated into the slaughterhouse, as is usually the case in Brazil (Sousa et al., 2017). Hence, no burdens were considered for this pathway in the upstream stage of S1. Finally, UCO was treated as an end-of-life product, *i.e.*, product at the end of its useful life that could potentially undergo reuse, recycling, or recovery (JCR, 2010). Therefore, no upstream burden was included.

On the other hand, System 2 (S2) treats the residual feedstock as a valuable product from the upstream stage, considering the increasing use and market for biomass residues. According to the JCR (2010), "if the market value of the waste/end-of-life product at its point of origin is above zero, in the LCA perspective it would be considered as a co-product, and the multifunctionality is to be solved by allocation." Likewise, the Roundtable on Sustainable Biomaterials (RSB, 2017) methodology uses this approach when the economic value of an output is greater than 5% of the total value of the other products generated in the same production process (RSB, 2017). This approach has also been adopted in some LCA studies for lignocellulosic ethanol (Bonomi et al., 2016) and RJF from tallow (Seber et al., 2014).

As cut-off criteria, the environmental burden related to the production and assembling of machinery and processing equipment, as well as building construction, was not included. Since the environmental impacts related to them are diluted over their lifetime, it is expected a relatively minor contribution to the results. Also, the environmental burden related to catalyst use was disregarded due to the lack of information on the production conditions and uncertainties regarding catalyst loads or lifetime.

2.1.2. Allocation procedures

The environmental burdens of each life cycle stage were partitioned among the multi-products as represented in Fig. 1, which is a more consistent approach for cause-oriented analyses, such as attributional studies (JCR, 2010; UNEP-SETAP, 2011). In this study, economic allocation was applied as a default method, *i.e.*, the partitioning was based on the market prices of each product. The allocation factors are presented in Supplementary material (Table SI.2), from the values informed in Table SI.1.

2.1.3. Land use change (LUC)

One of the motivations to use residual feedstocks for biofuel production is that, presumably, there would not be any additional land requirements. As a matter of fact, direct and indirect LUC (dLUC and iLUC) – which accounts for the carbon emissions from the conversion of the original land use and rebound effects in other locations, respectively – have been raised as a concern for biofuel production in general (Bailis and Baka, 2010; Han et al., 2017; Moreira et al., 2014; Staples et al., 2014).

Despite the relevant influence of the LUC on GHG emissions (Bailis and Baka, 2010; Han et al., 2017; Moreira et al., 2014; Staples et al., 2014), LUC impacts were not accounted for. Given the methodological approach used here (attributional LCA), the present study focuses on the environmental performance of each RJF pathway rather than evaluating the consequences outside of the system boundaries. Then iLUC would be out of the scope. In turn, dLUC was not also considered, since deforestation for the production of biofuels is very unlikely in Brazil due to the current legislation in the country (*e.g.*, Forest Code (BRAZIL, 2012) and RenovaBio (ANP, 2018)) as well as the international sustainability requirements on biofuels (*e.g.*, CORSIA (ICAO, 2019) and European's Renewable Energy Directive (European Parliament, 2009)). Nevertheless, the conversion of croplands and pasturelands may still lead to relevant carbon emissions or sequestration, which must be addressed on case by case basis.

2.1.4. Environmental impact categories

The life cycle impact assessment was performed according to the ReCiPe (H) midpoint method v.1.13 (Huijbregts et al., 2016) and included the following categories: climate change, terrestrial acidification, eutrophication, human and environmental toxicities, photochemical oxidant formation, particulate matter formation, and fossil depletion. Here, the results for eutrophication category correspond to the sum of freshwater and marine eutrophication values. Likewise, results for freshwater, marine, and terrestrial toxicity are combined in environmental toxicity category.

2.1.5. Database

The foreground systems were assembled using primary, secondary, and modeled data, as indicated in Section 2.2. For the background systems (*e.g.*, production of chemicals and utilities), inventories were taken from Ecoinvent v3.3 (Ecoinvent, 2016), USCLI (NREL, 2018), and GREET databases (ANL, 2018) and adapted to the Brazilian context whenever possible. SimaPro 8.3® (PRé-Sustainability, The Netherlands) was used as an auxiliary tool for the analysis.

2.2. Life cycle inventory (LCI)

2.2.1. Upstream stage

Among the oil-based pathways, the soybean production and harvesting conditions are fully described in Table SI.5, adapted from (IBICT/SICV, 2019) and based on 2012–2014 averaged data for Mato Grosso State, which is the major Brazilian producer (IBGE, 2019a).

The upstream stages for beef tallow production comprise the cattle management, slaughter, beef production, and rendering process. The full description of the LCI under Brazilian conditions was adapted from (Sousa et al., 2017) and available in Table SI.6. According to them, for simplification purposes, boneless meat and beef tallow are the only products considered at the slaughterhouse, while leather, edible offal, blood, and condemned parts were considered wastes.

The agricultural stage of the sugarcane-based pathways was described according to the *Virtual Sugarcane Biorefinery* (*VSB*) tool (LNBR, 2018) from averaged data of São Paulo State, which is the current major Brazilian producer (IBGE, 2019a). The *VSB* model covers the whole supply chain of Brazilian sugarcane with validated data. A complete mechanized harvesting process was assumed with 50% recovery of straw by bailing/loading systems and the agricultural use of vinasse and filter-cake returned from ethanol distillery (see Table SI.7). A general description and the main aspects found in VSB are presented in Bonomi et al. (2016).

The sugarcane residue-based pathways, *i.e.*, *via* 2G-ethanol and Fisher-Tropsch (FT) were modeled considering a mix of bagasse and straw as feedstock. This material is provided by an optimized 1G autonomous mill (Bonomi et al., 2016), which burns only the amount of biomass required to supply its steam demand. Hence, the upstream stage is composed of the sugarcane cultivation and harvesting and 1G-ethanol mill. A detailed LCI is presented in the Supplementary material (Table SI.8).

Finally, for the pathways involving wood residues, the upstream inventory was based on a Brazilian company that manufactures cellulose and paper from eucalyptus. The LCI represents the common practices for this crop (Coelho, 2018), which are listed in Table SI.9. The branches, top, and bark are chopped by a diesel-electric machine in a "full-tree" harvesting operation and transported to the plant.

2.2.2. Midstream stage

At this stage, only soybean extraction, UCO rendering, and the production of hydrated ethanol were considered. Soybean oil extraction using hexane was described by Sugawara (2012) and the corresponding LCI is provided in Table SI.10. The LCI for collecting and rendering UCO is based on Seber et al. (2014) (Table SI.11).

For the sugarcane-based pathways, *via* 1G ethanol, an optimized autonomous mill was considered, as represented by the *VSB* (Bonomi et al., 2016; LNBR, 2018) and adjusted to produce hydrated ethanol. A detailed LCI is in Table SI.12.

The 2G processes from sugarcane residues were modeled as standalone plants – *i.e.*, physically separated from the 1G process, to allow for an independent evaluation – considering two different technologies: enzymatic hydrolysis (2Gh) and gasification of lignocellulosic material with subsequent syngas fermentation (2Gs). The former is based on an advanced 2G technology, as described by the *VSB* models (Bonomi et al., 2016; LNBR, 2018), and further adjusted to produce hydrated ethanol. The *VSB* model considers that solid residues (*i.e.*, cellulignin) are used as an energy source in the Combined Heat and Power (CHP) system. The industrial effluents, such as vinasse and pre-treatment flash, could alternatively be used for biogas production, as suggested by Humbird et al. (2011). However, the presence of inhibitory agents, such as phenolic compounds, may cause difficulties in the biodigestion of stillage (España-Gamboa et al., 2011; Wilkie et al., 2000). On the other hand, this was not considered as an obstacle for its application on the field, as suggested by (LNBR, 2018). Detailed LCIs for sugarcane and wood residues are available in Tables SI.13 and SI.16, respectively. For wood residues, the 2Gh models in *VSB* were adapted to the composition presented in Table SI.3. Furthermore, the production process of the enzyme was based on Da Silva et al. (2014) considering the sugar input from an optimized annexed ethanol mill (Bonomi et al., 2016), as presented in Table SI.14.

The pathways from syngas fermentation were based on the models developed by de Medeiros et al. (2017) and adapted to the composition of both biomass sources, sugarcane residues and wood residues (for details, see Table SI.13). The process modeling considers steam generation by heat recovered from hot gases and power generation from unreacted syngas. The syngas fermentation parameters and liquid media composition are in line with those of Gaddy et al. (2007). The make-up media nutrients for syngas fermentation were simplified to account for the most relevant components, which are also available in (Ecoinvent, 2016). The wastewater leaving the process is assumed to undergo treatment before disposal or reuse, and the ashes from the gasification process are returned to the field to be used as fertilizers. Detailed LCIs are gathered in Table SI.15 for sugarcane residues and Table SI.17 for wood residues.

The main overall yields related to the upstream and midstream processes, for all pathways depicted in Fig. 1, are presented in Table SI.18.

2.2.3. Downstream stage

Three certified technologies, according to (ASTM, 2019), were considered for RJF production, whose LCIs were mostly based on the modeling performed by Klein et al. (2018), with some adaptations, as described below. A major difference from Klein et al. (2018) is that the hydrogen is supplied by an external plant (*i.e.*, the H_2 production system is outside of the system's boundaries), except for the FT process in which hydrogen is produced internally *via* gasification.

Here, the HEFA model considered the self-supply of utilities by the internal burning of light streams (*e.g.*, propane), which are produced at 102 kg/ton oil (Klein, 2019). This differs from Pearlson (2011) who reported external power and natural gas inputs and the light stream outputs. The airborne emissions were considered similar to the liquefied petroleum gases in an industrial boiler (ANL, 2018), assuming biogenic carbon. The wastewater undergoes treatment before disposal or reuse.

For the UCO pathway, the conversion performance was assumed to be similar to soybean oil in HEFA technology – as also assumed by Seber et al. (2014) and (de Jong et al., 2017) – because of the high consumption of soybean oil in Brazilian cuisine, *i.e.*, around 90% of vegetable oil consumed in 2008 (IBGE, 2019b). This assumption was deemed appropriate for the scope of this study, although the influence of UCO composition on HEFA yields should be further investigated. On the other hand, it is reasonable to suppose that the use of UCO for RJF production in large scales would not be feasible because of the constraints related to the logistics of its collection. In this sense, UCO is expected to be used as a co-feedstock with other oil-based materials, hence lowering the influence of its composition on the overall industrial yields.

For beef tallow, the hydrogen demand was adjusted according to Pearlson (2011) and Klein et al. (2018) and considering the different compositions of the feedstock (INRA, 2018). Utilities and conversion yields for the tallow-based pathway were kept the same as reported by Klein et al. (2018).

In ATJ technology, the steam demand is supplied by burning light hydrocarbons produced throughout the process (around 146 kg/ton ethanol), according to Klein et al. (2018) and Klein (2019). The wastewater was also assumed as properly treated without environmental burden to the reference flow.

Finally, the FT process was also based on Klein et al. (2018) and considered sugarcane residues and eucalyptus as feedstocks, on-site hydrogen production, and the use of light hydrocarbons (around 3.2 kg/ton lignocellulose material) as self-energy source. For practical purposes, the conversion yields from eucalyptus were assumed to be the same as eucalyptus residues. Wastewater treatment was also assumed as no additional environmental burdens to the system occur.

The overall yields and hydrogen input of the processes within RJF conversion processes are summarized in Table SI.19.

The LCI for hydrogen production was based on a Brazilian company (Marin, 2018), assuming steam methane reform (SMR) with a platinum catalyst. A detailed inventory can be found in Table SI.20.

2.2.4. Transportation and use

One-way distance was considered to evaluate transportation stage. In oil-based pathways, the distance between the soybean crop in Mato Grosso State to the extraction plant (Midstream) was set at 400 km. Collection and transportation of UCO to the rendering plant were set at 50 km, based on the average distance for recyclables collection by two cooperative units in a medium-sized city in Brazil (Lino et al., 2010). For sugarcane-based pathways, an average distance of 36 km was assumed to transport straw and stalks to the ethanol mill (LNBR, 2018) or FT plant. For wood-based pathways, this distance was defined as 40 km, which corresponds to the current economically feasible value to collect wood residues for use as an energy source (Coelho, 2018).

A default distance of midstream and slaughterhouse to downstream was set at 400 km. This considered possible values between a rendering plant, an extraction plant, or an ethanol distillery to the RJF plant, which was assumed to be near an oil refinery in São Paulo State.

Likewise, to supply the airport, 200 km was set for all pathways, which corresponds to the weighted distance between the three major Brazilian refineries of Jet A production — *i.e.*, REVAP (São Paulo State), REPLAN (São Paulo State), and REDUC (Rio de Janeiro State) (ANP, 2019) — to Guarulhos International Airport that is responsible for around 30% of kerosene consumption in Brazil (ANP, 2019). Specifically, for the FT pathways, with no midstream processes, a one-way distance of 600 km between the FT plant and the airport was assumed.

Transportation was considered to be entirely based on heavy trucks that meet the EURO4 emission standards (Ecoinvent, 2016). This inventory was adapted to the most commonly diesel consumed in Brazil and the current biodiesel blend (B10). Diesel S500, *i.e.*, with 500 ppm of sulfur content, corresponded to around 70% of the diesel consumed in Brazil in 2016, but the current efforts for S10 expansion is expected to decrease S500 contribution to 42% in 2026 (Coelho, 2017). For biodiesel, it was assumed that 82%, on average, of Brazilian biodiesel is derived from soybean oil and 18% from tallow (ANP, 2019). The inventories related to biodiesel production were reported by Sousa et al. (2017) and Sugawara (2012), while the airborne emissions from its use were adjusted considering: no sulfur, 20% increase of nitrogen dioxides, and decreases of 75%, 15%, and 40% for hydrocarbons, particulates, and carbon monoxide, respectively, as reported by the EPA (2002).

Finally, the emissions related to RJF use were assumed similar to a typical aircraft operation in an intracontinental trip, as reported by Ecoinvent (2016), with the following adjustments: reduction of 2% and 5% in carbon dioxide and nitrogen oxide emissions, respectively, due to lower heating, cetane number, and density of RJF in comparison with fossil kerosene; increase of 11% in water emissions; and no emissions of particulate matter and sulfur. The carbon emissions from RJF use were considered biogenic. These adjustments were made according to Moore et al. (2017), Stratton et al. (2011), Donohoo (2010), and Cavalett and Cherubini (2018) (Table SI.22).

2.2.5. Fossil kerosene (Jet A)

The fossil kerosene production was assumed to be similar to a typical oil refinery in the United States (NREL, 2018), as suggested by Sugawara (2012). The split of the multiple oil-products was adapted to the average production profile 2007–2017 of the three major Brazilian refineries: REVAP, REPLAN, and REDUC (ANP, 2019), which are responsible for around 40% of Brazilian oil products. The extraction of crude oil was taken from Ecoinvent Association (2016) and adapted to Brazilian conditions, as described in Table SI.23. The transportation of Jet A between refinery and airport was set in 200 km (one-way) by the same assumptions presented previously (Item 2.2.4).

2.3. Uncertainty and sensitivity analysis

The uncertainties and the significance of the results were assessed through a Monte Carlo analysis with 1000 trials. The parameter distributions related to the foreground systems were based on the original databases and adaptations from similar inventories in the literature. When data was not available, uncertainties were estimated according to the Pedigree Matrix (Goedkoop et al., 2016; Weidema and Wesnæs, 1996). All the assumptions and uncertainty data for the 44 parameters considered here for the foreground systems are indicated in Table SI.4A. For the background systems, it was assumed the uncertainty data already available on the Ecoinvent database (Ecoinvent, 2016).

Additionally, the sensitivity of the environmental trade-offs with respect to relevant parameters and methodological choices were evaluated as well. Conversion yields were varied according to the ranges reported in the literature (Table SI.4B). Given the relevance of the hydrogen supply for most pathways, the alternative route based on water electrolysis (WE) was also investigated, whose inventory is available in Table SI.24. The effect of different locations of the conversion plants with respect to the biomass sources and airports was assessed through a $\pm$ 50% allowance on transportation distances, except for the transportation of sugarcane and wood residues to the ethanol plant, which are already a well-established in the country (Bonomi et al., 2016; Coelho, 2018). As for the methodological choices, the effect of energy-based allocation (instead of economic allocation) was analyzed, following the parameters given in Table SI.2.

3. Results and discussion

This section is divided into five parts: in the first, the pathways are analyzed per impact category, considering the contribution of each life cycle stage; subsequently, the uncertainty of the previous results are discussed. The trade-offs between the climate change and the other impact categories are discussed in the third part; and in the fourth one, a sensitivity analysis is carried out. Finally, the environmental impacts estimated here are compared to other reports from the literature.

3.1. Environmental impacts assessment of RJF

In general, RJF from 1G pathways (*i.e.*, Soy oil/HEFA and SC_1G/ATJ) lead to higher impacts along its life cycle than 2G pathways at S1 (*i.e.*, waste-based pathways), mainly due to the environmental burden related to the upstream stage. On the other hand, the opposite is observed in some cases when the residual feedstock is assumed to be a valuable by-product (S2). The results are presented in Fig. 2. Table SI.26 in Supplementary material presents the contribution of each stage and related activities, which can support specific investigations.

Specifically at S2, the pathways based on sugarcane residues present higher values than wood residues-based ones in all impact categories. It is justified by the different system boundaries of wood and sugarcane residues (see Fig. 1), and different allocation factors (see Table SI.2). Furthermore, sugarcane crop presents a relative higher environmental impacts than wood crop. In other words, while at S1, the "upstream" of wood residues comprises only their collection and transport operations, no burden is allocated for sugarcane residues, which is assumed to be totally carried by ethanol. Otherwise, at S2, the wood residues take up 7.0% of the burden related to the eucalyptus crop by economic allocation and sugarcane residues bear 15% of the total burden estimated for the sugarcane crop and ethanol mill.

In turn, FT pathways tend to present the best environmental performance of all categories for the S1 and S2 approaches, even with the lowest overall yield (56 and 59 g RJF/kg feed$_{(db)}$ for sugarcane and wood residues, respectively) compared to other lignocellulosic-based processes, such as 77–59 g RJF/kg feed$_{(db)}$ for enzymatic hydrolysis (2Gh) and 71–64 g RJF/kg feed$_{(db)}$ for syngas fermentation (2Gs). FT pathways do not require a midstream stage and their downstream stage is self-supplied with hydrogen and utilities, which explains their environmental performance.

Regarding specific impact categories, around half of the GHG emissions related to RJFs from the 1G pathways (see Fig. 2.A) are associated with the upstream stage and can be explained by the combined effect of the use of nitrogen fertilizers (2.1% and 13.5% of the total GHG emissions for Soy oil/HEFA and SC_1G/ATJ, respectively), emissions from crops and industrial residues on the fields (22% and 9.8%), and diesel use in agricultural operations and input transportation (around 10% for both pathways). However, for 2G pathways when the upstream stage is taken into consideration (S2), GHG emissions reach 3.1 in WO/FT to 150 g CO_{2e}/MJ in tallow/HEFA. For the latter, the methane from the enteric digestion of cattle (4.6 kg CH_4/MJ_{RJF}) is responsible for around 70% of the GHG emissions assigned to the feedstock, even assuming a low allocation factor for tallow (3%) at the slaughterhouse gate.

The midstream stage is relevant in 2Gh pathways and corresponds to around half of the total GHG emissions at S1, *i.e.*, 11 g CO_{2e}/MJ. In this case, enzyme use, which demands natural gas and sugar for its production, is responsible for 25% of the total emissions of these pathways.

Despite similar ethanol yields, 2Gs pathways present lower values than 2Gh because of the low environmental burden related to the industrial inputs at the midstream stage, which corresponds to around 6.0% of the total GHG emissions (0.55 g CO_{2e}/MJ) (Tables S.16 and S.17). Even with the contribution of the industrial inputs reported by Handler et al. (2016) – but not detailed by them (*i.e.*, 1.30 g CO_{2e}/$MJ_{ethanol}$) – the GHG emissions of WO_2Gs/ATJ and SC_2Gs/ATJ would increase by 16% on average but would still be lower than the 2Gh pathways.

At the downstream stage, HEFA processes usually require three-to-four-fold more hydrogen in kg H_2/kg_feed (ANL, 2018; Klein et al., 2018; Pearlson, 2011) than ATJ technology (Han et al., 2017; Klein et al., 2018; Staples et al., 2014). Therefore, the GHG emissions related to hydrogen input in the HEFA pathways – *i.e.*, 7.4 g CO_{2e}/MJ for tallow and 8.8 g CO_{2e}/MJ for soybean and UCO – are in contrast to ATJ pathways, where the hydrogen input results in 4.6 g CO_{2e}/MJ. In general, hydrogen use contributes 15% (SC_1G/ATJ) and 23% (Soy oil/HEFA) of the total emissions in 1G pathways. For 2G pathways at S1, the contribution is around 20% for 2Gh-based pathways, 30–40% for 2Gs-based, and more than 60% for oil-based.

The fossil depletion category (Fig. 2.B) presents similar trends to those of the climate change results, except for Tallow/HEFA at S2, due to the biogenic methane emissions. In this category, hydrogen use is the main contributor. In HEFA-based pathways, this corresponds to 5.6. g_oil$_e$/MJ (soybean and UCO) and 4.7 g_oil$_e$/MJ for tallow, *i.e.*, more than 50% of the total environmental impact. In ATJ pathways, its contribution (2.9 g_oil$_e$/MJ) corresponds to 39% (SC_1G) to 68% (SC_2Gs at S1) of the total impact. Likewise, 1G pathways have a greater impact than the 2G ones in both approaches (S1 and S2), exclusively because of the upstream accounting. At the upstream stage, diesel use in agricultural operations, including inputs transportation, corresponds to around 20% of the total values in SC_1G and Soy oil/HEFA. At the midstream stage, around 20% of the total values in 2Gh pathways are related to ammonia input.

Terrestrial acidification is mostly related to NH_3, NO_x, and SO_x emissions, while eutrophication is related to the nutrient (nitrate and phosphorous) emissions into water bodies. Therefore, the relevant contribution of the upstream stage in 1G and 2G pathways at S2 (see Fig. 2.C and D) is, in general, mostly associated with the nitrogen input from chemical fertilizers and from organic substances (*e.g.*, industrial effluents or crop residues).

According to the LCA inventories (Tables SI.5 an SI.7), the total nitrogen input for sugarcane is lower (1.26 g N/MJ_{RJF}) than for soybean (1.88 g N/MJ_{RJF}). However, in sugarcane, over 60% of the nitrogen is obtained from chemical fertilizers (0.67 g N/MJ_{RJF}) and industrial residues (0.15 g N/MJ_{RJF}) (vinasse and filter cake); while in soybean crops, the major contributor is biological nitrogen fixation by the plant (1.76 g N/MJ_{RJF}). In this context, despite that the nitrate emissions are estimated from the total nitrogen input, ammonia emissions are estimated only from chemical fertilizers and manure (Nemecek and Kagi, 2007), which explains the higher values for SC_1G/ATJ than Soy oil/HEFA regarding terrestrial acidification and their similar values for eutrophication. Likewise, even 2G pathways from sugarcane residues at S2 present higher values than Soy oil/HEFA for terrestrial acidification. In turn, the considerable contribution from the upstream stage in Tallow/HEFA at S2 (around 90% of the total values) is mostly related to ammonia emitted from cattle urine, as reported by Seber et al. (2014).

For both categories, the contribution of the midstream stage in 2Gh pathways is mostly related to the enzyme input, which bears the impact of sugar production and corresponds to 20% and 30% of the total terrestrial acidification and eutrophication results at S1, respectively.

Regarding the eutrophication category, although some inventories (IBICT/SICV, 2019; Jungblut et al., 2007; Tsiropoulos et al., 2014) have

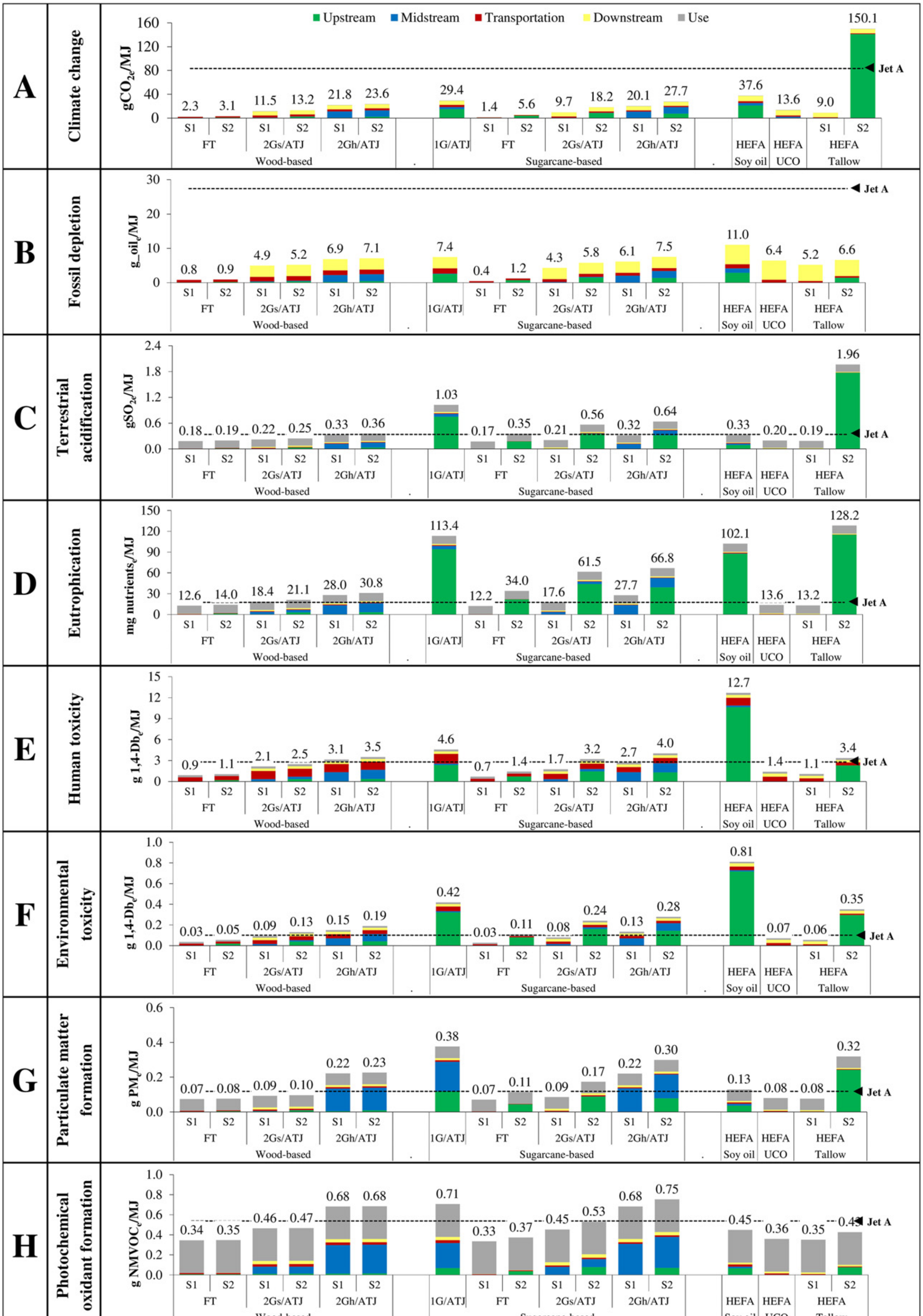

Fig. 2. LCA of RJF and Jet A (S1, residual feedstock as waste; S2, residual feedstock as by-product).

accounted for phosphorous emissions from fertilizers use based on general assumptions, none were considered here. As set out by Bonomi et al. (2016) and Cavalett et al. (2013), phosphorous leaching and loss by water erosion in Brazilian soil are not verified due to the high phosphorus-binding capacity of the soils and the flat landscape in the producing regions, which reduce this risk (Riskin et al., 2013).

Human and environmental toxicities are directly linked to the use of agrochemicals, including fertilizers and pesticides, at the upstream stage, which again explains the higher values of the 1G pathways than for the 2G pathways for both approaches (S1 and S2) (Fig. 2.E and F). In general, direct emissions from use of agrochemicals represent 11% and 50% of the environmental burden related to human toxicity in SC_1G/ATJ and Soy oil/HEFA, respectively; and less than 10% for residues-based pathways (S2). On the other hand, these emissions correspond to 60% of the environmental toxicity for 1G pathways, and 15 to 40% for 2G pathways (S2).

As stated by Macedo (2005), the more intense application of pesticides to soybean crops (estimated at 1.76 $kg/t_{soybean}$ or 96.9 mg/MJ_{RJF}) than sugarcane (0.02 $kg/t_{sugarcane}$ or 8.8 mg/MJ_{RJF}) confirms the considerable toxicity of Soy oil/HEFA. Likewise, the upstream accounting (S2) results in a significant increase in the values for pathways based on sugarcane residues and tallow, because of the allocated burden of the sugarcane crop and animal feed, respectively. Substantial variations for wood-based pathways between S1 and S2 are not seen due to the low allocation factor of wood residues compared to sugarcane residues and the relatively low use of pesticides (0.03 kg/t_{wood}).

The split of pesticides emissions to air/water/soil, which considerably influences toxicity impact categories, has been commonly simplified or omitted in several LCAs through the application of different arbitrary assumptions on splitting fractions (Berthoud et al., 2011; Bonomi et al., 2016; Jungblut et al., 2007; Nemecek and Kagi, 2007). Here, the split of pesticide emissions to soil, air, and water is assumed to be same for soybean, sugarcane, and wood – *i.e.*, 90% to soil; 9% to air, and 1% to water (Tables S.5, S.7, and S.9) – as suggested by the European Commision (2018). However, it is worth noting that different modeling options of pesticide emissions can influence the environmental assessment of agricultural products as concluded by Schmidt Rivera et al. (2017). On the same way, Nordborg (2013) reported a different split for pesticide emissions in Brazilian crops based on computational modeling, and considering different application techniques, climate conditions, and types of pesticides. In that study, for soybean, 0.4% of the pesticides would be emitted into air and 0.002% into surface water, for sugarcane, 10.5% would go into air and 0.4% into surface water. This discrepancy should be analyzed in future investigations.

The contribution of the transportation stage to human toxicity is related to brake wear emissions. They are relevant for SC_1G/ATJ (around 30% of the total environmental impact) and wood-based pathways (more 35% at S1), for which the transportation from field to ethanol mills was fully considered.

Particulate matter and photochemical oxidant formation are related to possible impacts on local air quality. For these categories, the burning of lignocellulosic material in the midstream stage of the SC_1G/ATJ (0.17 g PM_e/MJ) and 2Gh pathways (0.10 g PM_e/MJ) contributes with around 50% of particulate matter formation at S1 (Fig. 2.G). Likewise, process emissions (*e.g.*, ethanol releasing) contribute around 20% of the photochemical oxidant formation of these pathways (Fig. 2.G).

Specifically, for particulate matter formation, the contribution of the upstream stage in sugarcane-based pathways is mostly related to nitrogen oxide emissions from fertilizer use, *i.e.*, around of 25% (0.42 g NO_x/MJ_{RJF}) and 15% (0.17 g NO_x/MJ_{RJF}) of the total values in the SC_1G/ATJ and 2G pathways at S2, respectively. For Tallow/HEFA at S2, ammonia emissions from cattle urine (0.69 g NH_3/MJ_{RJF}) in the upstream stage are responsible for around 70% of the total environmental impact.

Regarding to photochemical oxidant formation, RJF use is responsible for, at least, 45% of the total results of each pathway (Fig. 2.H). However, it provides only 8% lower impact than those related to fossil fuel use for this category. According to the RJF use inventory (Table SI.22), while a large reduction in combustion-generated particles and low or no sulfur emissions are related to RJF use; no relevant reductions in carbon monoxide, hydrocarbons, and nitrogen oxides emissions – which influences this impact category – have been reported (Moore et al., 2017; Stratton et al., 2011). Nevertheless, Benosa et al. (2018) confirmed the benefits of alternative kerosene in reducing aviation emissions in the boundary layer (up to 1000 m). According to their report, the 50/50 blend of RJF and fossil kerosene provided lower sulfur dioxide emission and particulate matter impact on the ground-level than other strategies to improve air quality in airports, such as taxi out time reduction and ground support equipment electrification.

In general, in this study, assessment of the local impact, such as air quality, toxicity, acidification, and eutrophication was conducted by general characterization factors, which can be refined in future investigations considering a specific description of the region where the supply chain is to be implanted.

3.2. Uncertainty analysis

Considering the uncertainties related to the life cycle inventories, all base case values (deterministic results) presented in Fig. 2 are within the 95% confidence interval generated by the Monte Carlo analysis, *i.e.*, 2.5th percentile to 97.5th percentile (see "Base case" in Fig. 3). Furthermore, most of the base case values are near the median and mean values. Some discrepancies are observed when the upstream stage is accounted for, such as in SC_1G/ATJ, Soy oil/HEFA, and Tallow/HEFA (S2) for climate change and toxicity categories.

GHG emissions in the base case (see Fig. 2.A) are more optimistic than the median values from Monte Carlo simulations. While the base case reported 37.6 g CO_{2e}/MJ and 29.4 g CO_{2e}/MJ for Soy oil/HEFA and SC_1G/ATJ, respectively, the median emissions for these pathways are 42.6 g CO_{2e}/MJ (varying in 34.2 to 54.4 g CO_{2e}/MJ) and 32.6 g CO_{2e}/MJ (27.4–38.6 g CO_{2e}/MJ). In turn, the median emissions of Tallow/HEFA (S2) are 189 g CO_{2e}/MJ (146–521 g CO_{2e}/MJ) compared to 150 g CO_{2e}/MJ as reported in Fig. 2.A. The range related to N_2O emissions from fertilizers (IPCC, 2006) and CH_4 emissions from cattle management (Oliveira et al., 2016) are the main underlying reasons for this gap. Similarly, the uncertainty on pesticides application in soybean crop (Raucci et al., 2015) leads to median values for human and environmental toxicity of 14.4 g $1,4Db_e/MJ$ (10.3–22.8 g $1,4Db_e/MJ$) and 1.2 g $1,4Db_e/MJ$ (0.5–2.0 g $1,4Db_e/MJ$), respectively, while base case results are 12.7 and 0.8 g $1,4Db_e/MJ$ (see Fig. 2.E and F). All results of the Monte Carlo analysis are available in Table SI.27.

In addition, the uncertainty range of the results for each pathway can lead to no significant differences among them. Then, by Monte Carlo analysis, which was run in SimaPro 8.3®, it was possible to estimate the frequency when two compared pathways are different from each other during the trials. If the frequency of the difference is observed in more than 95% of the trials, it was assumed there is a significant difference among the pathways (Goedkoop et al., 2016). These comparisons are illustrated in Fig. SI.1.

For instance, the small difference observed between the results of eutrophication and environmental toxicity in SC_1G/ATJ and Soy oil/HEFA (see Fig. 2.D) are not considered significant, which means that during the trials Soy oil/HEFA can present higher values than SC_1G/ATJ, and *vice-versa*. Likewise, the differences between Soy oil/HEFA and Tallow/HEFA (S2) are not significant for eutrophication and photochemical oxidant formation.

Finally, wood-based pathways in comparison with sugarcane residues at S1 are significantly different only for Fischer-Tropsch (FT). On the other hand, when the upstream is accounted for, *i.e.*, in S2, the sugarcane residues-based pathways are significantly higher than wood-based pathways in climate change, terrestrial acidification, eutrophication, and particulate matter formation.

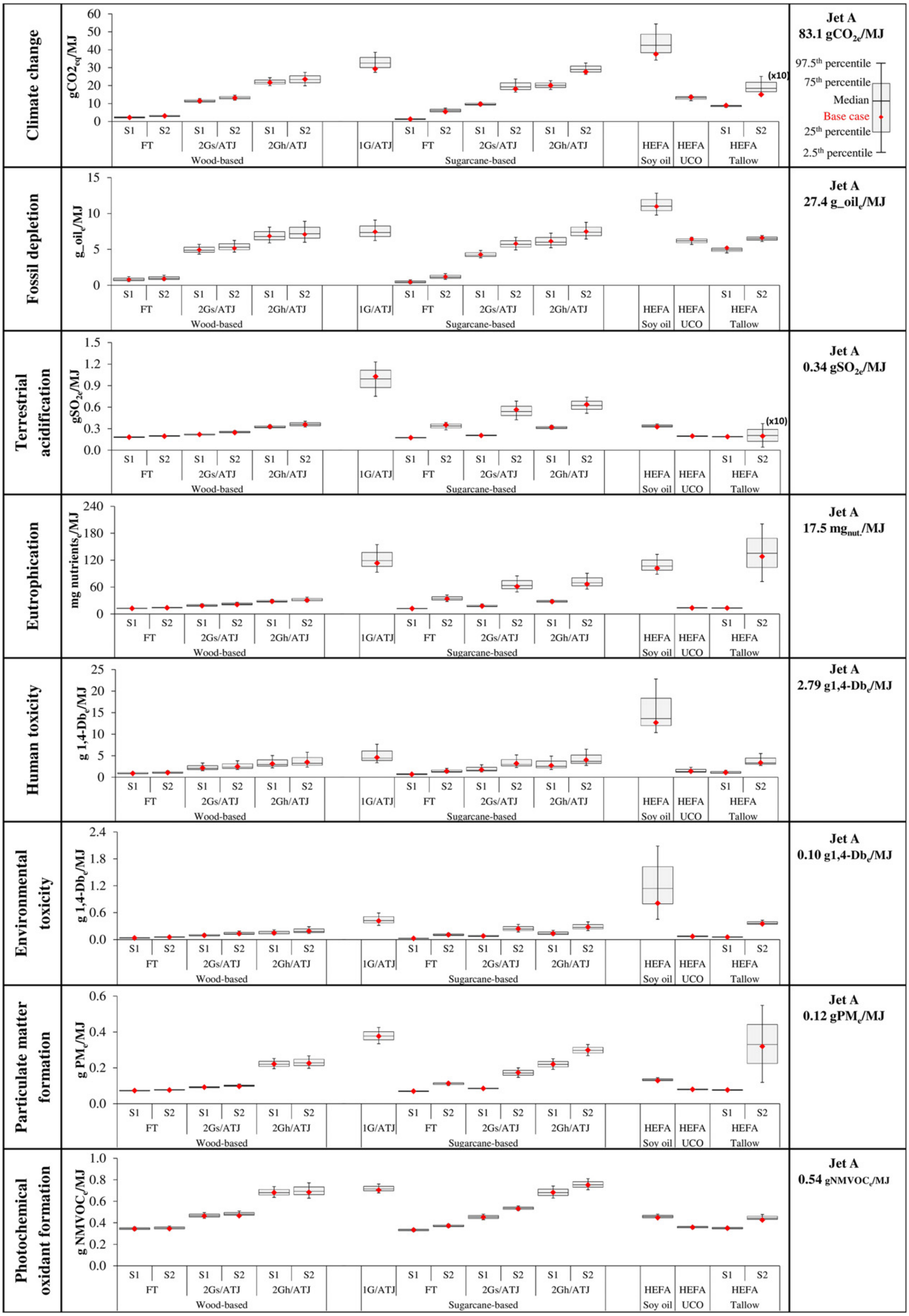

Fig. 3. Uncertainty analysis of LCA of RJF (S1, feedstock as waste; S2, feedstock as by-product). Climate change and terrestrial acidification results for Tallow/HEFA (S2) were adjusted to better fit to the graph scale.

3.3. Environmental trade-offs

All pathways reported a possible reduction in GHG emissions compared to fossil kerosene (Jet A), but this does not occur in the other impact categories (see Fig. 4). By the deterministic results (base case values), 1G pathways – *i.e.*, Soy oil/HEFA and SC_1G/ATJ – provide a GHG reduction of 55% and 65%, respectively, compared to Jet A. However, they present relevant values for local impacts. For example, the Soy oil/HEFA reports human toxicity impacts three-fold higher than those for the sugarcane-based pathway and around five-fold higher than those for Jet A (Fig. 4.A), mainly due to agrochemicals use. On the other hand, the SC_1G/ATJ pathway (Fig. 4.D) presents two-fold higher terrestrial acidification impacts than for soybean-based (and six-fold higher than Jet A. Similarly, higher particulate matter and photochemical oxidant formation impacts (around three-fold and 30% higher, respectively, than Jet A) are seen for SC_1G/ATJ. In turn, the results related to eutrophication for Soy oil/HEFA and SC_1G/ATJ, which are not significantly different from each other, are around six-fold higher than Jet A.

Some of these trade-offs are discussed by Cox et al. (2014), who reported low GHG emissions and fossil fuel dependency for the sugarcane-based pathway and high values for eutrophication and water consumption. Similarly, Klein et al. (2018) highlighted the benefits of RJF produced in integrated sugarcane biorefineries for global-scale impact categories, such as climate change and fossil depletion, which contrasted with high local impact (human toxicity, terrestrial acidification, and agricultural land occupation), mostly observed at the agricultural stage.

When residual feedstock is treated as waste (S1), some trade-offs are observed but only in 2Gh pathways (Fig. 4.E and G). While these pathways provide a GHG reduction of 74% (WO_2Gh) to 76% (SC_2Gh), photochemical oxidant and particulate matter formation impacts are 30% and 90% higher than Jet A, on average, respectively. Pathways based on wood residues lead to slightly higher environmental impacts than those obtained for sugarcane residues in all assessed categories at S1, and this may be explained by the difference in ethanol production yield and the boundaries of the LCA, as mentioned in Section 3.1. However, as mentioned previously, these differences are significant only when FT technology is considered.

Furthermore, no trade-offs are observed for the other pathways at S1, whose potential GHG reduction is estimated around 97% for the FT pathways, 89% for Tallow/HEFA, 86% for 2Gs/ATJ pathways, and 84% for UCO/HEFA. These pathways lead to the fewest environmental impacts, following this order, for all categories except fossil depletion, in which Jet A presents the highest values compared with all pathways in both approaches (S1 and S2).

On the other hand, when the residual feedstock is treated as a by-product (S2), relevant trade-offs take place in 2G pathways. For

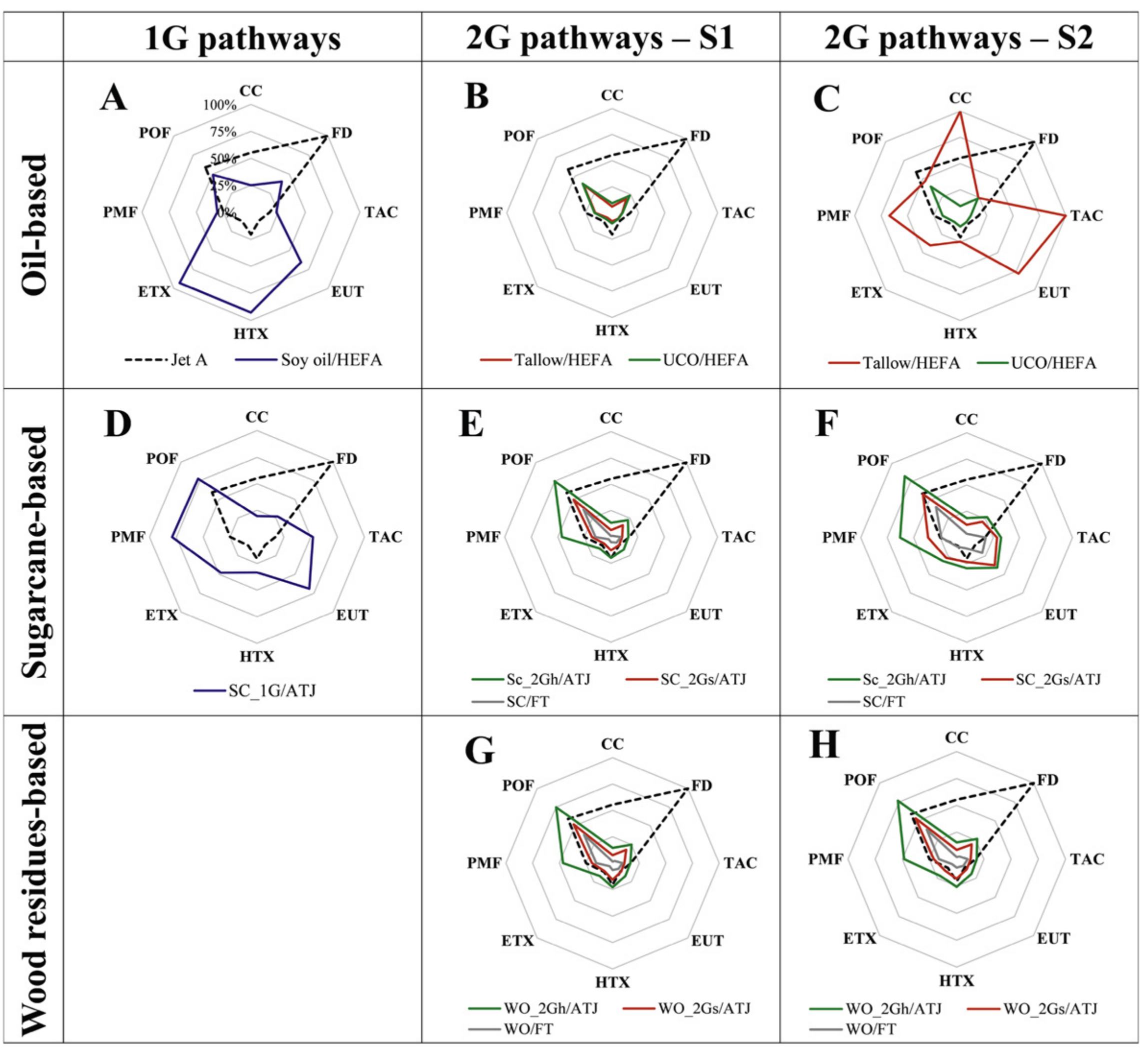

Fig. 4. Environmental trade-offs of RJF pathways normalized by the highest values in each impact category according to the deterministic results (base case values) presented in Fig. 2. CC, climate change; FD, fossil depletion; TAC, terrestrial acidification; EUT, eutrophication; HTX, human toxicity; ETX, environmental toxicity; PMF, particulate material formation; POF, photochemical oxidant formation. S1, residual feedstock as waste; S2, residual feedstock as by-product.

pathways based on sugarcane residues (Fig. 4.F), while providing a GHG reduction of 67% (SC_2Gh) to 78% (SC_2Gs), terrestrial acidification and eutrophication become, on average, 77% and four-fold higher than Jet A, respectively. The aspects related to sugarcane crop inventory, such as nitrogen use, and the high allocation factor applied to sugarcane residues at the upstream stage explain these values.

Otherwise, no relevant differences are observed for wood-based pathways between S1 (Fig. 4.G) and S2 (Fig. 4.H). With a potential GHG reduction of 72% (WO_2Gh) and 84% (WO_2Gs), the results in S2 are significantly different from S1 only for some categories, such as terrestrial acidification and eutrophication.

At S2, the largest discrepancy compared to the values estimated in S1 is observed for Tallow/HEFA (Fig. 4.C), which confirms the high impacts related to pasture activities. For climate change, the values become 80% higher than Jet A in base case, or 120% higher (median value) by Monte Carlo analysis. Even compared to 1G pathways (Fig. 4.A and D), the results for terrestrial acidification and eutrophication are 90% and 12% higher than SC_1G/ATJ, respectively.

3.4. Sensitivity analysis

In general, the results of terrestrial acidification, eutrophication, and toxicity range in the same order as the upstream yield variations. In turn, climate change, fossil depletion, and toxicity impacts vary similarly to the range of the downstream yields. Hydrogen production *via* water electrolysis (WE) would imply increasing GHG emissions of up to 13% in Soy oil/HEFA (4.7 g CO_{2e}/MJ), as well as decreasing fossil depletion in 30% of the same pathway (Fig. 5). Pathways based on sugarcane residues at S2 are considerably sensitive to the energy allocation method. They would present higher impacts even than the 1G pathways, and trade-offs would be observed even in FT pathway. The sensitivity analysis is detailed in Supplementary material (Fig. SI.2).

3.5. Comparison with other studies

The GHG emissions related to the RJF life cycle are the primary impact category discussed in the literature. In this context, comparing the results achieved here to those reported in other studies can help to identify trends and differences. In the case of the Soy oil/HEFA pathway, the results are similar to those published by Han et al. (2013), who considered soybean production in the United States by energy allocation, and within the range of other oil-bearing feedstocks, such as jatropha, rapeseed, camelina, and palm (Fig. 6). According to the authors, the main differences between Soy oil/HEFA and the other oil-based pathways are explained by the high fertilizer consumption of jatropha, camelina, and rapeseed crops and the high palm oil yield. Camelina oil as feedstock was also studied by Li and Mupondwa (2014) under five endpoints environmental impacts. Different designs for HEFA process and different demand of fertilizers and crop yields also explain the range of the results reported by them. The direct comparison of their results for climate change to those reported here would not be correct, because they accounted credits to the co-products, while here, in an attributional approach, it was not assumed.

For UCO and tallow, the results described here are lower than those published in other studies mainly because of inventory aspects and system boundaries. For example, in the tallow inventory, Seber et al. (2014) considered the rendering process separately to the slaughter process. Likewise, when tallow is assumed to be a by-product of meat production, the discrepancy between the results estimated here (148 g CO_{2e}/MJ) with respect to those from Seber et al. (2014) (87 g CO_{2e}/MJ) is explained mainly by the estimations of methane emissions during the animal's lifespan. According to the pastoral system of beef production (assumed here), around 174 kg CH_4/cattle head are emitted along the three years required for the cattle to reach a weight of 450 kg (Sousa et al., 2017). Seber et al. (2014) is based on a feedlot system, in which, 57 kg CH_4/cattle head are emitted along the 1.5 years required for the cattle to be ready for slaughter.

Other differences among the oil-based pathways derive from the design of RJF conversion technology: while other studies (de Jong et al., 2017; Han et al., 2013; Pearlson, 2011; Seber et al., 2014) assumed external utilities' demand and light hydrocarbons production, such as propane, the internal use of this light stream was considered here, with power surplus generation. As mentioned previously (Section 2.2.3), the HEFA process modeled by Klein et al. (2018) – and adopted here – aims to assure the self-supply of utilities, which commonly result in good performance from an LCA perspective. However, an economic assessment will indicate the best design of a HEFA plant.

In the case of ATJ pathways, Klein et al. (2018) considered an integrated plant (to the ethanol mill) with on-site hydrogen production from water electrolysis. This leads to lower emissions. In turn, for SC_1G/ATJ, de Jong et al. (2017) assumed lower values for chemical fertilizer input for the sugarcane crop (0.80 kg N/ton sugarcane) than those considered here (1.26 kg N/ton sugarcane). The higher GHG emissions for Corn/ATJ (de Jong et al., 2017) than those for the sugarcane-based pathway can be explained by the significant nitrogen (15 kg N/ton corn) and diesel demand (5 L/ton corn) at the upstream stage, added to the overall performance of the conversion process, which also accounted for external utilities.

For the residue-based pathways, such as corn stover, de Jong et al. (2017) and Han et al. (2017) considered additional fertilizer demand as nutrient compensation due to crop residue removal (around 30 kg NPK/ton corn stover), which explains the difference between their results and those presented here. Cavalett and Cherubini (2018) reported slightly higher values for RJF from forest residues in Norway, due, most

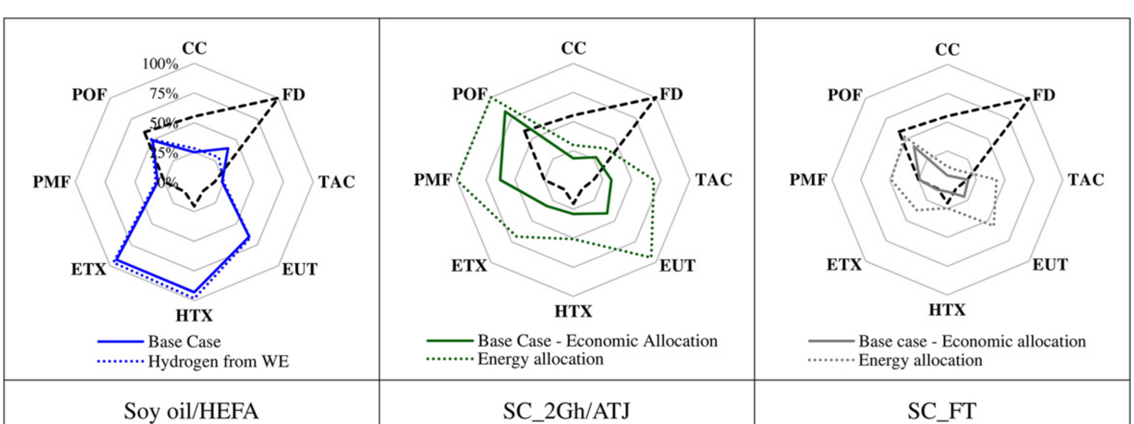

Fig. 5. Sensitive analysis for some key parameters. S2, residual feedstock as by-product.

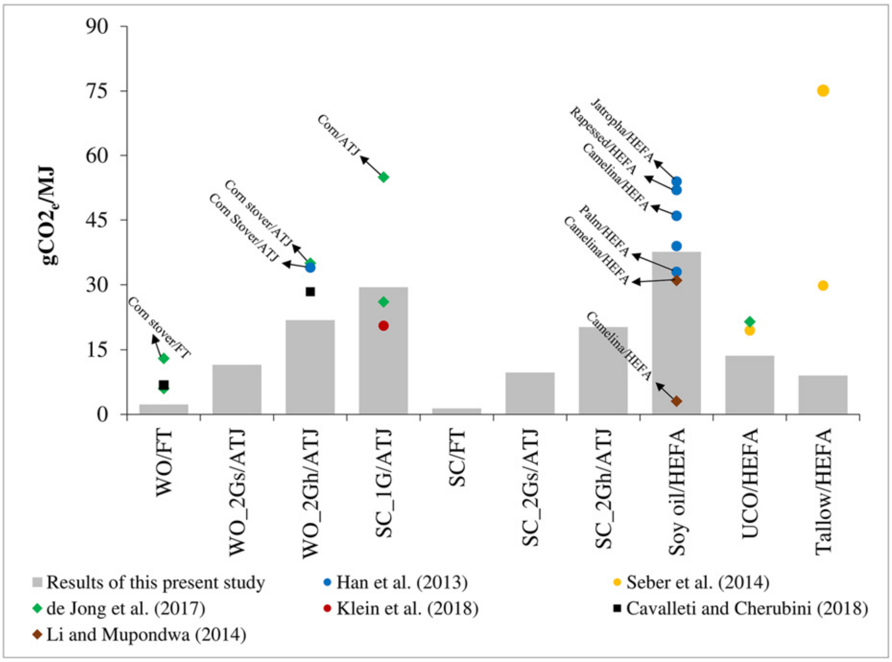

Fig. 6. GHG emissions for RJF production and use in comparison with other studies (Cavalett and Cherubini, 2018; de Jong et al., 2017; Han et al., 2013; Klein et al., 2018; Li and Mupondwa, 2014; Staples et al., 2014); residues-based pathways by S1 approach; none LUC aspects were considered; dots without label indicate results for the same pathway (feedstock and conversion technologies) as here analyzed.

likely, to different agricultural inputs, transportation distances, and operations (*e.g.*, harvesting, chipping, and processing) for forest residues.

Other impact categories are briefly discussed in some studies. According to Klein et al. (2018), the performance of RJF from 1G ethanol for terrestrial acidification is around two-fold higher than the fossil kerosene, which is similar to what is estimated here. On the other hand, relative to Jet A, RJF provides less fossil depletion (−85% *vs.* −73% in this study) and human toxicity (27% *vs.* 64% in this study), due to inventory aspects, such as on-site hydrogen production by water electrolysis.

Regarding pathways from wood-residues, Cavalett and Cherubini (2018) recommended the FT pathway as the most interesting option in terms of environmental performance. However, in contrast to what is estimated here, they reported higher impact in some categories compared to Jet A: terrestrial acidification (−24% *vs.* −46% in this present study), particulate matter formation (−11% *vs.* −38% in this study), and photochemical oxidant formation (−6% *vs.* −36% in this study). For these same categories, those authors reported that RJF from 2G ethanol provided greater impact relative to Jet A, such as terrestrial acidification (13% *vs.* −35% in this study), particulate material (34% *vs.* −22% in this study), and photochemical oxidant formation (30% *vs.* −14% in this study). The description of the whole supply chain in Norway – which included field and industrial operations, transportation, and RJF use – can explain these differences.

4. Conclusions

An attributional life cycle assessment of 10 different pathways to produce RJF in Brazil was carried out in the present study. Potential 1G pathways from soybean and sugarcane and residue-based pathways, *i.e.*, 2G pathways from wood, sugarcane, UCO, and tallow, were evaluated through eight impact categories.

In general, while RJF pathways provide lower global-scale impact than fossil kerosene (Jet A), such as climate change and fossil depletion, relevant trade-offs are observed in categories related to local impacts, such as eutrophication, toxicity and air quality-related categories. The 1G pathways have potential to provide a GHG emission reduction of over 50% with respect to fossil kerosene (Jet A), even considering the uncertainties related to the life cycle inventories. However, sugarcane-based pathway (SC_1G/ATJ) is related to high impacts in terms of eutrophication and air quality, mostly because of fertilizer use and bagasse burning at the ethanol mill. Furthermore, the soybean-based pathway (Soy oil/HEFA) causes large impacts on human and environmental toxicity, because of agrochemical applications. The GHG emission reductions are estimated to be around 70% in the 2G pathways, when the residual feedstock is treated as waste and, consequently, the environmental burden of the upstream stage is not considered. In these cases, no relevant trade-offs are observed, except for air quality impacts observed in hydrolysis-based pathways with wood and sugarcane residues, due to biomass burning at the ethanol mill.

However, when treating residual feedstocks as by-products, the environmental performance of some pathways changes considerably and relevant trade-offs take place. For instance, the beef tallow pathway (Tallow/HEFA) leads to 80% higher GHG emissions than Jet A, as well as larger impacts regarding terrestrial acidification and eutrophication than 1G pathways. Similarly, pathways based on sugarcane residues, although providing a potential GHG reduction of 67% (SC_2Gh/ATJ) to 94% (SC/FT), feature higher impacts than Soy oil/HEFA for terrestrial acidification, particulate matter, and photochemical oxidant formation. In this context, wood-based pathways perform better than sugarcane residues, due to the relatively low environmental burden of the upstream stage allocated to this feedstock.

The definition of what is considered waste (or not), as already observed in low carbon policies, can support (or not) the use of residues for biofuel production. Nevertheless, several of these residual materials have been used in specific markets and are treated as valuable products by their sector. This study does not intend to advocate for a specific pathway, but, rather, indicates what values could be achieved for different impact categories depending on how the feedstock is treated in the LCA.

Pathways with low dependency on industrial inputs featured the best performances. Then, FT pathways in both approaches, followed by syngas fermentation-based ones, represent high potential reduction in GHG emissions (over 75%) with no relevant environmental trade-offs. UCO/HEFA is also an interesting option, but the considerable demand for hydrogen poses some limitations. Further, the effective potential of the feedstock supply and maturity of these technologies can be obstacles to their quick start-up.

It must be noted that the findings of the present analysis are based inventories that reflect the conditions of Brazilian agriculture and the forecasted performances of promising RJFs production routes. As such, the results obtained here cannot be simply extrapolated to other scopes given the relevance of the upstream stages for 1G pathways. Nevertheless, future analyses may benefit from the detailed life cycle inventories assembled in this work, whereas the findings for residues-based pathways tend to be less sensitive to the geographical scope.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.136696.

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