Full length article

Environmental life cycle assessment of polypropylene made from used cooking oil

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A B S T R A C T

Used cooking oil (UCO) has received much attention as feedstock for the production of renewable fuels and bio-based materials. This study aims to assess the environmental impact of UCO-based polypropylene (PP) by a cradle-to-factory gate Life-Cycle Assessment (LCA). 16 impact categories were assessed. The results were interpreted with normalization and weighting steps. For several multi-output processes, different allocation procedures were scrutinized. On a normalized and weighted basis, the environmental impacts of UCO-based PP are dominated by climate change (28%), fossil resource use (23%) and water use (11%). The following environmental hotspots are identified: the polymerization process (38%), the production of hydrogen (21%), the production of LPG (18%) and the combustion of LPG (8%). Compared to petrochemical PP, cradle to factory gate impact reductions of 40–62% for climate change and 80–86% for fossil fuel resource use can be achieved by UCO-based PP, depending on the allocation approach chosen. For other impact categories, the environmental footprint of bio-based PP is strongly influenced by the choice of the allocation method.

1. Introduction

It was 1954 when Natta and his research group first polymerized propylene to a crystalline isotactic polypropylene (PP) (Raos, 2019). Already at that time, Natta realized that the properties of polypropylene could have introduced new trends in the world of plastics (Natta, 1955). Nowadays, PP represents one of the most widely used plastics in Europe (nearly 20% of the total consumed plastics) (PlasticsEurope, 2017). In 2018, the global production of polypropylene resin was 56 Mt. The demand is projected to increase to 88 Mt by 2026 (Reports and Data, 2019). The high demand for polypropylene is due to its versatility, which allows its use in many applications such as food packaging, construction pipes, and automotive parts. The properties that especially make polypropylene multipurpose are associated with its high melting point, low density, excellent stiffness and strength (PlasticsEurope, 2014a).

In the last decade, due to the growing demand for plastics and related concerns about their climate change impact, bio-based plastics have attracted attention as a possible option to replace petrochemical plastics. Many studies have highlighted that bio-based plastics could potentially offer a lower carbon footprint (Shen et al., 2010; Spierling et al., 2018; Suwanmanee et al., 2013; Vera et al., 2019; Weiss et al., 2012). However, their environmental performance depends on the type of polymer, the impact category in focus, the selected system boundaries, the type of biomass

feedstock and its final application and the supply chain (Choi et al., 2018; Ingrao, Gigli, and Siracusa, 2017; van der Harst, Potting, and Kroeze, 2014; Weiss et al., 2012). In 2017, the global production capacity of bio-based plastic materials reached 2.05 Mt and was expected to increase to 2.44 Mt by 2022 (European Bioplastics, 2017). In particular, bio-based PP entered the market in 2019 with a production capacity of about 19 kt, which is predicted to increase by about six folds by 2024 (European Bioplastics, 2019). The three main synthesis routes for bio-based PP are: 1) using bio-ethanol from sugar fermentation (Kikuchi, Oshita, Mayumi, and Hirao, 2017; Machado, Walter, and Cunha, 2016; Niaounakis, 2015), 2) using bio-syngas (Gay, Pope, and Wharton, 2011; Kikuchi et al., 2017) and 3) using hydrotreatment of used cooking oil (UCO) (Neste, 2018).

To our knowledge, only two peer-reviewed environmental life cycle assessments (LCAs) of bio-based PP are publicly available (Kikuchi et al., 2017; Mayumi, Kikuchi, and Hirao, 2010), and no peer-reviewed LCA has been conducted for the third route, i.e. UCO-based PP. Mayumi et al. performed a cradle-to-factory gate LCA of biomass-derived PP and polyethylene (PE) at the design stage (Mayumi et al., 2010). In the study, they quantified greenhouse gas (GHG) emissions of polyolefins made from the waste wood-syngas route. They found out that bio-based PP and PE could lead to higher GHG emissions compared to the petrochemical counterparts. The impacts of the bio-based polymers were dominated by biomass production and conversion processes.

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Kikuchi et al. (2017) investigated PP and PE made from sugarcane-ethanol and woody-biomass-derived syngas based on data from demonstration plants and simulations. Defining one liter of ethanol as functional unit, they found that the highest GHG reduction can be obtained by using ethanol for PE production (a saving of about 50% compared to petrochemical PE), followed by use of ethanol as transport fuel (a saving of about 40% compared to gasoline), and PP production (a saving of about 15–20%). They concluded that PP production from bio-syngas offers relatively limited GHG emission reduction.

This study aims to investigate the environmental impacts of bio-based PP obtained through the third route, i.e., via hydrotreatment of UCO. UCO is a waste and thus does not compete with food and feed. It also avoids potential land use changes. In the EU-28, the total potential UCO available from the gastronomy sector, food processors and households is estimated at around 4 Mt per year (EUBIA, 2020). These features make it an attractive feedstock for future transportation fuels and material production (Patel et al., 2017; Tsoutsos, Tournaki, Paraíba, and Kaminaris, 2016). Primary data were collected from Neste Oyj based on a new commercial production facility in Europe (Neste, 2019).

The primary objective of this study is to provide a full picture of the environmental footprint of UCO-PP. Unlike the previously published LCAs of bio-based PP, which focused on climate change only, this study assesses 16 mid-point environmental impact categories following the recommendation of impact category selection by the European Commission's Product Environmental Footprint Category Rules (PEFCR) guidance (draft version 6.3) (European Commission, 2018). Such a wide selection of indicators has been rarely reported for the environmental assessment of bio-based chemicals and products (Broeren et al., 2017; Spierling et al., 2018). For bio-based materials, environmental indicators such as land use, eutrophication, and acidification should not be ignored before investment or a policy decision is made (Broeren et al., 2017; Weiss et al., 2012). Since there are several multifunctional processes involved, the second objective of the study is to gain insights into how the allocation choices could influence the LCA results. Despite that LCA is a standardized methodology, multifunctionality is one of the main remaining issues in LCA (Reap, Roman, Duncan, and Bras, 2008). The findings of this LCA should be used both to understand the full environmental impacts, originated from both resources and emissions, and also to provide recommendations for future EU policy directives on the allocation choices for innovative bio-based plastics. The policy-level allocation recommendation is so far only available for renewable transport fuels and bioenergy (European Commission, 2016). The insights gained from this case study could help to reveal some complexity and demonstrate the influence of the allocation decisions in the LCAs of innovative bio-based products.

2. Material and methods

2.1. Goal and scope definition

Life Cycle Assessment (LCA) is a widely applied standardized method used to assess the environmental impacts of a product or a service. In particular, this LCA study focuses on UCO-based PP and has been developed within the EU Bio-SPRI[1] project. The LCA was conducted according to ISO 14040 and ISO 14044. The draft PEFCR guidance was used as practical guidance when the guidance from the ISO standards were insufficient (European Commission, 2018; ISO, 2006a, ISO, 2006b). The main goal of this LCA is to assess the impact of UCO-based PP to identify the environmental hotspots of the life cycle. The second goal of this LCA is to add further valuable research to the open debate of solving multifunctionality of biorefineries (as detailed in

section *Multifunctionality*). The results of this study are intended to be used by the industry for further process improvement, policy makers and the LCA community.

Based on the goal of the study, the functional unit (FU) is defined as 1 kg of polypropylene. A cradle-to-factory gate scope and an attributional approach are adopted in the LCA. Fig. 1 shows the process diagram of the production of UCO-based PP. Used cooking oil is converted into high value hydrotreated vegetable oil (HVO) by Neste NEXBTL technology. Together with the HVO renewable diesel grade product, a renewable HVO naphtha grade product is obtained from the hydrotreatment process. This study focuses on the cracking of this "bio-naphtha" to obtain propylene via a process equivalent to petrochemical steam cracking.

Accordingly to the location of Neste Oyj's biorefinery (Rotterdam), the geographic scope is defined as the Netherlands. This reflects the specific situation of UCO collected from the Netherlands and nearby regions and all the major processes for the conversion into polypropylene occurring in the Netherlands as well with the exception of steam cracking occurring in a neighbor country. Nevertheless, when a specific inventory for the Netherlands was not available, or a specific process occurs in another EU country, average European data have been used.

The temporal scope is current (2018) to the near future (5–10 years), and the technological scope is defined as the status-quo technology which is ready for commercialization (technology readiness level 8, based on definition reported in (European Commission, 2015)).

Sixteen mid-point impact categories are selected to analyze the full environmental footprint. The adopted impact assessment models for each impact category are listed in Table 1. Their selection is based on the recommendation of PEFCR draft guidance (version 6.3), which was the version available at the time when the study was conducted. Differently, from the PEFCR guidance, particulate matter (PM) and land use are assessed using the methods recommended by the PEF guide (European Commission, 2012).

Due to the many impact categories considered, normalization and weighting are applied to identify the *overall* environmental hotspots. To ensure that the same impact assessment models are used for characterization and normalization, the following selection has been done. For water use and resource depletion categories, the normalization factors are retrieved from PEFCR guidance v.6.3. For all the other impact categories, per capita, EU 27 normalization factors (2010) are retrieved from ILCD 2015 (Benini et al., 2014). Table 1 reports the normalization and weighting factors applied in this study.

2.2. Life cycle inventory modeling

2.2.1. Unit processes, data, and assumptions

For the foreground system, primary data were collected from Neste. Those are site-specific data. The background data were largely based on the Ecoinvent database (version 3.4) and PlasticsEurope's Eco-profiles (PlasticsEurope, 2014a). For unavailable data, assumptions were made based on literature and/or validated by Neste. Important assumptions have been scrutinized by sensitivity analyses in the discussion section (*Data uncertainty*).

The production of UCO-based PP starts from the collection of UCO. In the baseline calculations, UCO has been considered as waste and, therefore, entering the system "free of burdens" (cut-off approach). A sensitivity analysis of this approach can be found in the discussion section (*Model uncertainty: multifunctionality*). The impacts of the collection stage are taken into account in the LCA. UCO is sourced from restaurants and commercial buildings mainly in the nearby regions of the bio-refinery country of location. The collection of UCO from restaurants and other users to the biorefinery is carried out by trucks and by water and distances assumed based on Neste's specific supply chain.

During the NEXBTL process, the oil is pre-treated and deoxygenized under high pressure to transportation fuel quality using hydrogen. The

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[1] BIOSPRI (Bioeconomy: Support to Policy for Research and Innovation) project funded by DG RTD of the European Commission.
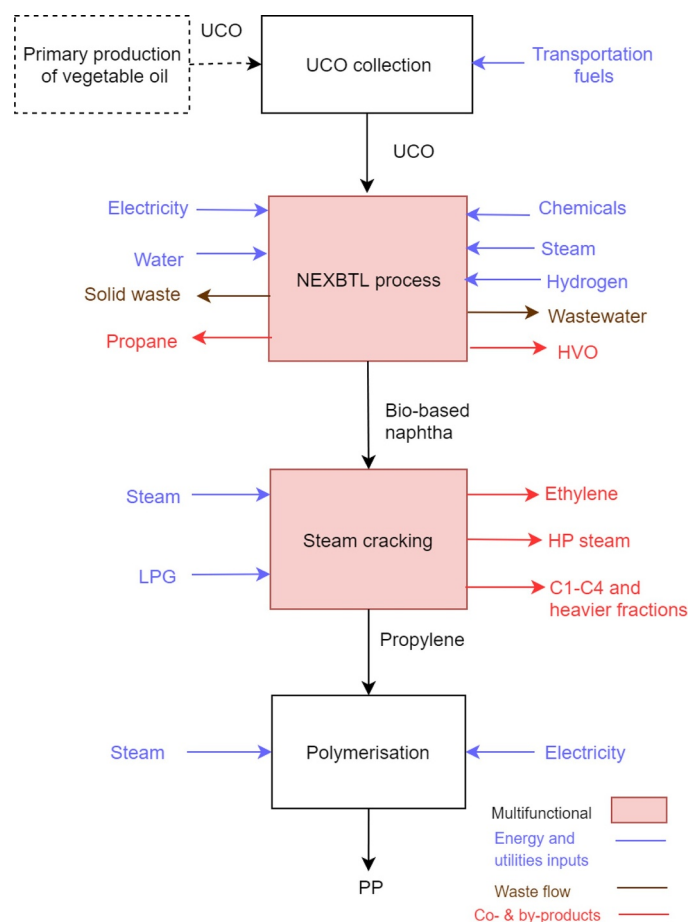
Fig. 1. Flow diagram of the production of polypropylene from used cooking oil (UCO). The dashed box represents the primary production of vegetable oil, which is a unit process that is out from the system boundaries in the baseline analysis.

triglycerides of UCO are converted to saturated straight and branched-chain hydrocarbons and oxygen (of the triglycerides eliminated). In the chemical reaction shown by Eq. (1), a triglyceride with formula $C_{57}H_{104}O_6$ is taken as an example. Hydrogen is produced by steam reforming of natural gas and fuel gas.

$$C_{57}H_{104}O_6 + 15\,H_2 \rightarrow 3\,C_{18}H_{38} + C_3H_8 + 6\,H_2O \qquad (1)$$
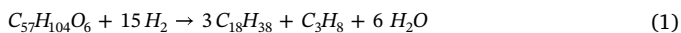
The main inventory data related to the NEXBTL unit process are collected in Table 2. For each ton of pre-treated oil, 1.02 t of UCO are needed (Nikander S., 2008). Steam is produced in the refinery while the Dutch national grid supplies the electricity. The amounts of steam reported in the inventory tables are equivalent kilograms, recalculated to keep into account the different energy content between the actual steam flows and the chosen dataset (equivalent kilograms on an energy basis). The chosen dataset represents the production of 1 kg of steam used for heating in the chemical industry. The heat is produced with the average fuel mix used in the European chemical industry (Althaus et al., 2007). The process releases wastewater that is treated on-site.

After hydrotreatment, bio-based naphtha is transported by train[2] to the steam cracking unit (distance based on Neste's specific case). During steam cracking, bio-based naphtha is diluted with steam and cracked into smaller hydrocarbons such as propylene and ethylene. The reaction is highly endothermic; therefore the feed is heated in a furnace burning fuel gas. The mass and energy inputs of the steam cracking unit in real operation are kept confidential and therefore the process has been modeled based on literature and is not Neste- specific. Process data

from Karimzadeh, Godini, and Ghashghaee, 2009 were modified considering the same feed capacity as the real operating pilot. The direct emissions released by the combustion of propane and butane are retrieved from the US Environmental Protection Agency dataset for industrial boilers (US EPA, 2008). In the calculations, the mix of 50% propane and 50% butane is assumed according to the dataset used for the production of LPG (Büsser, 2010). A selection of the main inventory data of this unit process is shown in Table 3.

The last process is polymerization of the propylene to obtain PP. The polymerization of bio-based propylene is identical to that of petrochemical propylene. The polymerization process has been extrapolated from the most recent PlasticsEurope's Eco-profiles (PlasticsEurope, 2014a) as follows: assuming that 1.02 kg (based on (Joosten, 1998)) of propylene are required to produce 1 kg of polypropylene, the impact of the polymerization process has been obtained by subtraction. The PlasticsEurope's Ecoprofile (PlasticsEurope, 2014b) has been widely used as benchmarks for comparison. However, they are often regarded as "black box" data because of a lack of transparency constrained by confidentiality.

As a consequence, it is challenging to interpret the impacts of the polymers fully. This can be considered a limitation of the study because of the restricted interpretation of the polymerization process due to the use of this dataset (see section *Data uncertainty*). Nevertheless, polymerization has also been modeled according to the Matter (MATerials Technologies for CO_2 Emission Reduction) study of 1998 for sensitivity analysis, and when necessary the comparison has been used as a validation test (Joosten, 1998). Despite the Matter study is twenty years old, it has been selected since it is the only transparent dataset for PP polymerization available in the public domain.

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[2] From Ecoinvent 3.4: Transport , freight train {Europe without Switzerland}| market for | APOS

Table 1
Selected impact categories and models, normalization and weighting factors.

Impact Category	Unit	Impact assessment models	Normalization factors EU 27 per person (Benini et al., 2014; European Commission, 2018)	Weighting factors with toxicity (Benini et al., 2014; European Commission, 2018)
Climate change	kg CO_2eq	IPCC 2013, GWP 100a with carbon climate feedback (Hartmann, Tank, and Rusticucci, 2013)	9.22E+03	21.06
Ozone depletion	kg CFC-11 eq	(Ramanathan and Feng, 2009)	2.16E-02	6.31
Human toxicity, non-cancer effects	CTUh	USEtox (recommended + interim) (Rosenbaum et al., 2008)	5.33E-04	1.84
Human toxicity, cancer effects	CTUh	USEtox (recommended + interim) (Rosenbaum et al., 2008)	3.69E-05	2.13
Particulate matter	kg PM2.5 eq	(Rabl, Spadaro, and Holland, 2014)	3.80E+00	8.96
Ionizing radiation Human Health (HH)	kBq U235 eq	(Frischknecht, Braunschweig, Hofstetter, and Suter, 2000)	1.13E+03	5.01
Photochemical ozone formation	kg NMVOC eq	(van Zelm et al., 2008)	3.17E+01	4.78
Acidification	molc $H+$ eq	(Posch et al., 2008)	4.73E+01	6.20
Terrestrial eutrophication	molc N eq	(Posch et al., 2008)	1.76E+02	3.71
Freshwater eutrophication	kg P eq	(Goedkoop et al., 2009)	1.48E+01	2.80
Marine eutrophication	kg N eq	(Goedkoop et al., 2009)	1.69E+01	2.96
Freshwater ecotoxicity	CTUe	USEtox (recommended + interim) (Rosenbaum et al., 2008)	8.74E+03	1.92
Land transformation	kg C deficit	Soil Organic Matter model (Milà et al., 2007)	7.48E+04	7.94
Water use	m3	AWARE factors (Boulay et al., 2015)	1.15E+04	8.51
Resource use, minerals and metals	kg Sb eq	(van Oers, de Koning, Guinée, and Huppes, 2002)	5.79E-02	7.55
Resource use, fossil fuels	MJ	(van Oers et al., 2002)	6.53E+04	8.32

2.2.2. Multifunctionality

According to ISO 14044, multifunctionality (or commonly also referred to as ''allocation'') should be solved using the following hierarchy (ISO, 2006b):

1) Avoiding allocation by subdivision or system expansion (i.e., expanding the product system to include the additional functions related to the co-products);
2) Allocation underlying physical relationships (i.e., an allocation that reflects how the inputs and outputs are changed by quantitative changes in the products of the system); and
3) Allocation (partitioning) based on other relationships (e.g., economic allocation).

In this context, the framework followed in this LCA to identify the impact of the choice of allocation approach is detailed. In the product system studied, there are two processes where more than one useful product is delivered: hydrotreatment and steam cracking (see Fig. 1).

The hydrotreatment process mainly delivers three products: hydrotreated vegetable oil (renewable diesel), propane and bio-based naphtha whose percentages are not disclosed. In this study, bio-based naphtha is the precursor of propylene while the other two co-products are sold. In this LCA, the problem of how to assign the environmental impact to the multiple products of hydrotreatment has been solved through energy allocation. Concerning the hierarchy above, the allocation is not avoided due to the inapplicability of subdivision (the process cannot be further subdivided). System expansion, both enlargement and reduction approaches, is also not possible. In fact, according to the goal and scope of the study, the functional unit cannot be enlarged to include all the co-functions. Concerning the inapplicability of system reduction, bio-based naphtha is a non-dominant by-product and therefore, the criterion of physical significance is not respected (i.e., the mass ratio of bio-naphtha is in the magnitude of a few percents to that of HVO, see Table 2). Allocation by physical causality is not applied because this would require a mathematical model (commonly based on linear programming) that is not available (Azapagic and Clift, 1999; Mackenzie, Leinonen, and Kyriazakis, 2017; Moretti, Moro, Edwards, Rocco, and Colombo, 2017). Among the possible remaining allocation methods, energy has been chosen according to RED recommendations when dealing with transportation fuels (European Commission, 2009; European Commission, 2016). Although naphtha is not used as transportation fuels, it is a by-product of fuel production (HVO) and therefore, RED recommendations are followed to respect the energy balances.

Nevertheless, it should be noticed that the RED focuses only on greenhouse gas emissions while 16 impact categories are analyzed in this study. For this reason, the authors have performed a sensitivity analysis on all possible allocation approaches used in the study (see the discussion Section 4.2). Energy allocation has been adopted for the hydrotreatment process.

The steam cracking unit delivers two main products, i.e., propylene and ethylene, other cracked gasses and steam (see Fig. 1 and Table 3). Propylene is in the focus of this study. Nevertheless, ethylene represents the biggest mass fraction among the cracked gasses. The steam cracking process also delivers 2.2 kg (6 MJ) of net industrial steam per kg propylene. Similar to the hydrotreatment unit process, subdivision, system expansion, and physical causality are not applicable. Avoiding allocation by substituting all propylene co- and by-products is not possible because a non-dominant product is in focus. For this unit process, a hybrid method has been chosen. Steam is directly used for other processes of the same biorefinery, and, otherwise, should be produced as marginal production of refinery steam from Ecoinvent (Steam, in chemical industry {RER}| production | Conseq). Direct substitution has been therefore applied to the net production of high pressure (HP) steam as, in this case, it may represent physical causality better than other arbitrary allocations (as highlighted by the PEFCR draft

Table 2

Input and output data per 1 kg of bio-based naphtha from UCO (NEXBTL unit process-hydrotreatment plus pre-treatment).

Flow	Data	Process	References on which foreground data are based on (and background data when different from Ecoivent)
Inputs			
Collected UCO	49.0 kg		Nikander, 2008
Phosphoric acid	28.0 g	Phosphoric acid, industrial grade, without water, in 85% solution state {RER}| purification of wet-process phosphoric acid to industrial grade, product in 85% solution state | APOS	(Edwards et al., 2017)
Process chemicals	0.1 kg	Chemical, inorganic {GLO}| production | APOS	Nikander, 2008
Water	5.0 kg	Water, decarbonised, at user {RER}| water production and supply, decarbonised | APOS	(Edwards et al., 2017; Nikander S., 2008)
Sodium hydroxide	48.0 g	Sodium hydroxide, chlor-alkali production mix, at plant/RER (PlasticsEurope. Industry data 2.0 project)	(Edwards et al., 2017; Fröhlich et al., 2013)
Electricity	6.0 MJ	Electricity, medium voltage {NL}| market group for | APOS	(Edwards et al., 2017; Nikander S., 2008)
Steam	10.0 kg	Steam, in chemical industry {RER}| production | APOS	(Edwards et al., 2017; Nikander S., 2008)
Hydrogen	1.7 kg	Hydrogen (reformer) E (PlasticsEurope. Industry data 2.0 project)	(Edwards et al., 2017; Nikander S., 2008; PlasticsEurope, 2005)
Nitrogen	1.6 g	Nitrogen, liquid {RER}| market for | APOS	(Edwards et al., 2017; Nikander S., 2008)
Outputs			
Bio-based naphtha,	1.0 kg		Along with HVO diesel and propane whose percentages are not disclosed.
Wastes			
Wastewater (output to technosphere: waste to treatment)	8.8 L	Wastewater, average {Europe without Switzerland}| market for wastewater, average | APOS	(Edwards et al., 2017; Nikander S., 2008)
Solid waste going to incineration	0.5 kg	Final waste flow, waste to incineration	Nikander, 2008

Table 3

Selected inventory data of the steam cracking process per 1 kg of bio-based propylene made from bio-based Naphtha.

Flow	Foreground data	Adopted background processes from Ecoinvent or modeled direct emissions	References on which these data are based on
Inputs			
Bio-based Naphtha	2.67 kg		Naphtha from hydrotreatment. (Karimzadeh et al., 2009)
LPG	0.63 kg	Liquefied petroleum gas {CH}| market for | APOS	(Karimzadeh et al., 2009; US EPA, 2008). Notice that only 0.43 kg of this LPG is burnt while the rest pumped with the feed.
Steam	2.9 kg		(Karimzadeh et al., 2009). This steam is the sum of diluted steam (66%) and boiling feed water (34%).
Outputs			
Propylene	1.0 kg		
Steam	5.1 kg	Steam, in chemical industry {RER}| production | Conseq (dataset used for net production)	(Karimzadeh et al., 2009) Steam conditions: 520 °C and 112 bar
Other cracked gasses and heavier products	Not disclosed		Ethylene is the major co-product and is accompanied by several other by-products (e.g. hydrogen, methane, benzene) whose percentages are not disclosed.
Direct Emissions			
Nitrogen oxides	1.3E-03 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
Carbon dioxide of fossil origins	1.3E+00 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
Carbon monoxide	7.5E-04 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
Methane	1.9E-05 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
Dinitrogen monoxide	8.5E-05 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
Particulate < 2.5 um	5.7E-05 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
Particulates, >2.5 μm, and <10 μm	1.9E-05 kg	Emissions to air due to LPG burning	(Karimzadeh et al., 2009; US EPA, 2008).
voc, volatile organic compounds	2.5e-05 kg	emissions to air due to lpg burning	(Karimzadeh et al., 2009; US EPA, 2008).
Sulfur oxides	1.6e-06 kg	emissions to air due to lpg burning	(Karimzadeh et al., 2009; US EPA, 2008).

guidance). Energy allocation has been applied to all the other co-(by-) products resulting in 20% of the remaining impacts allocated to propylene.

2.2.3. Biogenic carbon accounting

The carbon content of UCO-based PP originates from the CO_2 sequestered by biomass. According to the PEFCR draft guidance, only biogenic carbon emitted later than 100 years after its absorption shall be considered permanent storage (European Commission, 2018). Permanent storage results in a carbon credit to be assigned to the bio-based product. When biogenic carbon is emitted earlier than 100 years, no carbon credits must be assigned for temporally carbon storage or delayed emissions (European Commission, 2018). In particular, the PEFCR guidance recommends to not assign carbon credits for cradle-to-user assessments (European Commission, 2018). Nevertheless, the

biogenic carbon content at the factory gate "shall be reported as additional information" (European Commission, 2018). Accordingly, the authors have therefore proceeded as follows: the climate change impact is reported both with and without biogenic carbon removals while only the second one has been considered for the weighted results. Considering the chemical formula of propylene C_3H_6, per kg PP, the biogenic carbon removal corresponds to 3.14 kg CO_2eq.

3. Results

3.1. Impact assessment and interpretation

The cradle-to-factory gate environmental impact results are shown in Table 4, while the breakdown results of each unit process are illustrated in Fig. 2 (see Appendix A for numerical values behind the figure).

Table 4

Cradle-to-factory gate environmental impacts of 1 kg UCO-based PP.

Impact Category	Unit	Value	Normalized and weighted scores (Total 100%)
Climate change (without biogenic carbon removal (BCR))	kg CO$_2$eq	0.63	28%
Climate change, with biogenic carbon removal (BCR)	kg CO2eq	−2.51	Not applicable
Ozone depletion	kg CFC-11 eq	9.0E-08	1%
Human toxicity, non-cancer effects	CTUh	1.1E-08	1%
Human toxicity, cancer effects	CTUh	5.5E-09	6%
Particulate matter	kg PM2.5 eq	1.2E-04	5%
Ionizing radiation HH	kBq U235 eq	6.4E-02	6%
Photochemical ozone formation	kg NMVOC eq	1.9E-03	6%
Acidification	molc H+ eq	2.1E-03	5%
Terrestrial eutrophication	molc N eq	6.0E-03	2%
Freshwater eutrophication	kg P eq	8.7E-06	0%
Marine eutrophication	kg N eq	5.6E-04	2%
Freshwater ecotoxicity	CTUe	2.2E-01	1%
Land transformation	kg C deficit	1.1	2%
Water use	m3	7.4E-01	11%
Resource use, minerals and metals	kg Sb eq	3.2E-07	1%
Resource use, fossil fuels	MJ	9.30	23%
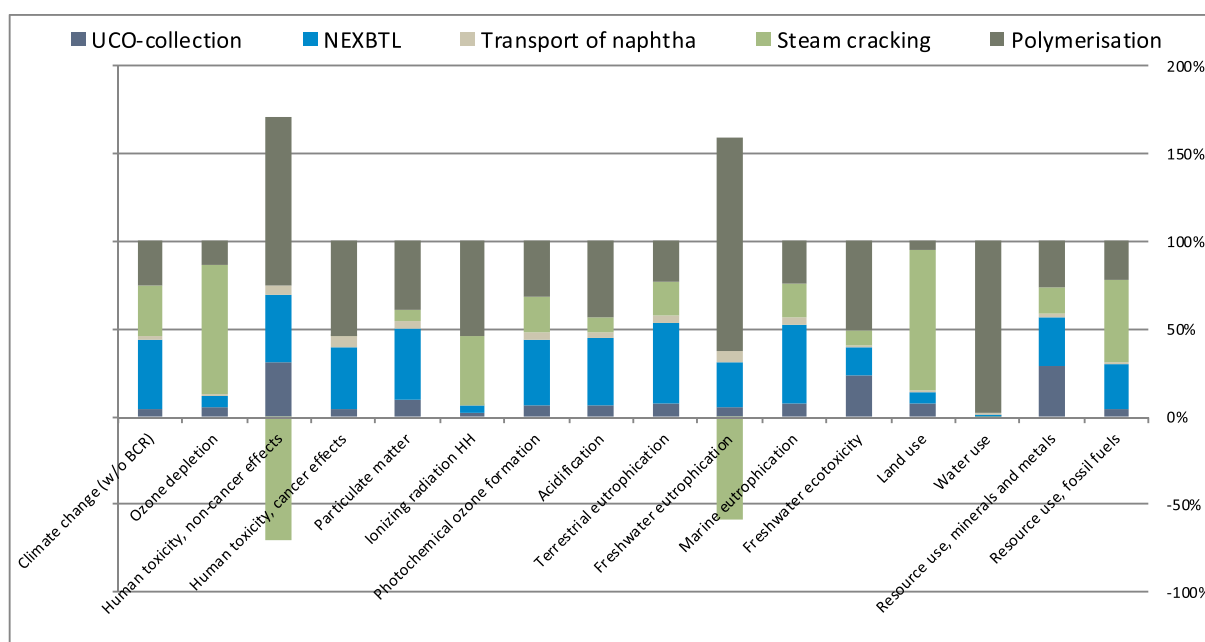
Fig. 2. Breakdown of cradle-to-factory gate environmental impact of UCO-based PP per key-unit process (climate change shown without biogenic carbon removal (BCR)).

From Table 4, it can be seen that climate change (28%), fossil resource use (23%) and water use (11%) as the most important environmental impacts of UCO-based PP. Fig. 1 shows that the NEXBTL process (hydrotreatment plus pre-treatment), steam cracking and polymerization are the three most significant key-unit processes in terms of environmental impact. On a weighted basis, polymerization contributes 38%, steam cracking 26% and hydrotreatment 23% of the total cradle-to-factory gate impacts.[3] The impact is almost entirely caused by the production (18%) and combustion of LPG (8%) and the production of hydrogen (21%) respectively for steam cracking and hydrotreatment (environmental hotspots). In particular, 7% out of 8% share of LPG combustion is caused by releasing GHG emissions. UCO collection and transportation of naphtha account only for 5% and 2%

respectively. Pre-treatment represents the remaining 6%.

The hydrotreatment process has a significant contribution (20–40%) in the following impact categories (see Fig. 2): climate change, human toxicity without cancer effects, human toxicity with cancer effects, particulate matter, photochemical ozone formation, acidification, terrestrial eutrophication, marine eutrophication, and resource use categories. For almost all the above impact categories, hydrogen production from steam reforming is the most (88–97%) relevant source of environmental impacts of the hydrotreatment process. The only exception is human toxicity without cancer effects, where most of the impact is due to producing electricity (37%) and phosphoric acid (31%).

Since a PlasticsEurope's "black box" Ecoprofile has been used for polymerization (see Section 2.2.1), an interpretation at the activity level is not possible. Nevertheless, it is possible to identify the five impact categories where polymerization contributes with the highest share (on a weighted basis) on the total impact (see Appendix A): water use (10%), climate change (7%), resource use of fossil fuels (5%), human toxicity (cancer) (3%) and ionizing radiation (3%).

From Fig. 2, it is possible to see that steam cracking shows negative

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[3] These shares of impact highlighted by the weighted results are not affected by the choice of applying weighting with toxicity instead of without toxicity. Indeed, changing weighting factors, only the shares of steam cracking and polymerization vary to 28% and 36% respectively.

impacts in two categories: *human toxicity without cancer effects* and *freshwater eutrophication*. The ''credit'' received from the substitution of the net steam produced during the steam cracking overcompensates the two impacts caused by LPG production and combustion. The steam cracking process is responsible for a significant share of environmental impact in several impact categories: namely, climate change (ca. 30%), ozone depletion (ca. 75%), ionizing radiation HH (40%), land use (80%) and fossil fuels resource use (ca.50%). In particular, LPG production and its combustion account respectively for 18% and 82% of the cradle-to-factory gate impact on climate change. The production of LPG alone is instead entirely responsible for the impact caused in the other four impact categories.

4. Discussion

4.1. Data uncertainty

This section reports several sensitivity analyses related to data uncertainty. The first assumption that is discussed is related to UCO collection. UCO was assumed to be sourced locally in the nearby regions of the biorefinery, based on Neste's specific case. This assumption led to a small overall impact (5%) from UCO collection. Nevertheless, UCO has attracted much attention as one of the bio-based feedstocks to achieve European renewable energy and greenhouse gas (GHG) emissions targets. This has led to an increase in demand and the international trade of UCO (De Mora, Torres, and Valero, 2015). Accordingly, it is reasonable that in the case of larger scale production of bio-based PP, UCO may be collected globally. Globally sourced UCO has been modeled considering the macro-areas, which together account for more than 90% of the 2017 Dutch consumption: Asia (40%), US (25%), West EU (20%) and Netherlands (15%) (Nederlandse Emissieautoriteit, 2017). According to a market study by (Toop et al., 2013), China has the highest UCO collectable potential. We assume that UCO is shipped[4] from China (port Shanghai) to represent the imported Asian UCO. For the US and Western Europe, New York and London have been considered for calculating the transportation distances, while the collection in the Netherlands has been modeled assuming the same distances used for the baseline calculations. The first column of Table 5 shows the changes in the results of environmental impact results when UCO is globally sourced. Compared to the baseline analysis, the change of UCO sources leads to a significant increase in all 16 impacts. The most affected categories are particulate matter (+60%), photochemical ozone formation (+48%), acidification (+78%), terrestrial and marine eutrophication (+55–57%) and human toxicity without cancer effect (+22%). Overall, on a weighted basis, the environmental impact increases by 19%.

Another important assumption is related to the dataset used for hydrogen production, which is among the main environmental hotspots of this route. In the investigated system, hydrogen is produced from steam reforming of natural gas. According to the selected dataset (PlasticsEurope, 2005), the production of 1 kg of hydrogen generates 9.4 kg CO_2eq. Nevertheless, literature reports GHG emissions from steam reforming of natural gas ranging from 8.9 to 12.9 kg CO_2 eq./ kgH_2 (Bhandari, Trudewind, and Zap, 2012). Accordingly, the climate change impact of bio-based PP production could vary in the range from −2% to 12% considering other datasets. In the future, environmental impact reductions could be achieved if hydrogen could be produced using renewable energy, i.e. via electrolysis powered by photovoltaics (Fernández-Dacosta, Shen, Schakel, Ramirez, and Kramer, 2019). In such a case, the climate change impact of UCO-PP could potentially be reduced by a third. However, as green hydrogen is not expected to be commercially viable within the next decade, it is out of scope for our LCA, and therefore not investigated in more detail.

LPG production and combustion has been identified as the second environmental hotspot of this route. The composition of LPG is variable depending on the location where it is sourced. For example, it can be 25% propane/75% butane in Italy and 95%/5% in Sweden (Saleh, 2008). In the baseline, we assumed that petrochemical LPG is used to produce the UCO-based PP. However, it is possible to optimize the process energy requirement by using the propane from UCO obtained from the hydrotreatment to meet the need of LPG for dilution of the steam cracking and by using the methane produced in the steam cracking as energy source (see Fig. 1). The environmental impact of UCO-based PP would vary as shown in the third column of Table 5. The overall environmental footprint of UCO-based PP would be reduced by 34%. In particular, these reductions of impact are high for climate change (−42%), ozone depletion (−86%), human toxicity without cancer effects (−44%), particulate matter (−31%), ionizing radiation HH (−41%), land use (−95%) and fossil resources (−64%). It is possible to notice that these reductions are even higher than the percentage of the impact caused by steam cracking on the overall environmental impact. This is consistent with the substitution approach used for steam produced. In fact, for these impact categories, the impact caused by steam cracking becomes negative when renewable propane and methane are used instead of LPG. The reason is that the impact caused by the production and combustion of renewable propane and methane (not the allocation of impact to methane produced by steam cracking) becomes lower than the credit for steam production (substitution).

Another important assumption is related to the datasets selected to model polymerization. As highlighted in the previous section, the impact of polymerization is particularly significant in five impact categories (on a weighted basis). For these impact categories, the share on the total impact has been validated using the data from the Matter study (Joosten, 1998). According to the Matter study, PP polymerization requires 2.1 MJ of electricity[5] and 1.3 MJ of steam[6] (averaged[7]) per kg of polypropylene. Using the inventory data from Matter study instead of PlasticsEurope (see section *Life cycle inventory modeling*), the share of polymerization would shift from 96% to 88% for water use, from 25% to 46% for climate change, from 22% to 40% for resource use of fossil fuels, from 54% to 48% from human toxicity (cancer) and from 54% to 59% for ionizing radiation. Overall, considering all the impact categories, the total share of polymerization would shift from 38% to 48% on a weighted basis. The authors consider the 9% difference in line with the different temporal scope of the two datasets (due to improvements in process efficiencies that have occurred over 20 years).

Fig. 3 summarises the results of the sensitivity analyses on data uncertainty on weighted bases. The variations are shown using the baseline values as 100%.

4.2. Model uncertainty: multifunctionality

This section provides a sensitivity analysis of the allocation approaches that were selected for the baseline calculations. The first sensitivity is related to UCO, which has been considered as a waste, being its use promoted in the European Union (European Commission, 2016). Accordingly, UCO has been treated with a cut-off approach in the baseline calculations. Nevertheless, the increasing demand for UCO for renewable diesel production has driven the high price of UCO in the past decade (De Mora et al., 2015). Accordingly, it might be argued that UCO should be considered as a by-product rather than a waste and a part of the impact caused in the first life (e.g., vegetable oil production) should be assigned to the recycled function (e.g., PP). For this

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[4] From Ecoinvent 3.4: Transport, freight, sea, transoceanic ship {GLO}| market for | APOS)

[5] Dataset used from Ecoinvent 3.4: Electricity, medium voltage {RER}| market group for | APOS

[6] Dataset used from Ecoinvent 3.4: Steam, in chemical industry {RER}| production | APOS

[7] Ranging between 0.8 and 1.8 MJ depending if the polymerization occurs in liquid phase, gas phase or suspension

Table 5
Variation of cradle-to-factory gate impact assessment results by different assumptions on UCO collection and process optimization of using renewable propane and methane.

Impact Category	Increase of impact (%) shifting from UCO locally sourced to sourced globally	Increase of impact (%) (negative values stand for a decrease) changing from conventional LPG in the baseline to the renewable propane and methane scenario
Climate change	10%	−42%
Ozone depletion	11%	−86%
Human toxicity, non-cancer effects	22%	−44%
Human toxicity, cancer effects	7%	2%
Particulate matter	60%	−31%
Ionizing radiation HH	6%	−41%
Photochemical ozone formation	48%	−5%
Acidification	78%	−17%
Terrestrial eutrophication	57%	4%
Freshwater eutrophication	12%	−15%
Marine eutrophication	55%	4%
Freshwater ecotoxicity	6%	−12%
Land transformation	13%	−95%
Water use	1%	0%
Resource use, minerals and metals	4%	2%
Resource use, fossil fuels	10%	−64%
Total weighted results	19%	−34%

Table 6
Variation of cradle-to-factory gate impact assessment results by using 50/50 method on UCO open-loop recycling.

Impact Category	Increase (%) (negative values stand for a decrease) 50/50 Method palm oil	Increase (%) (negative values stand for a decrease) 50/50 method soybean oil
Climate change	17%	58%
Ozone depletion	6%	28%
Human toxicity, non-cancer effects	−12%	−3701%
Human toxicity, cancer effects	16%	166%
Particulate matter	113%	485%
Ionizing radiation HH	4%	12%
Photochemical ozone formation	42%	164%
Acidification	37%	127%
Terrestrial eutrophication	40%	155%
Freshwater eutrophication	37%	1282%
Marine eutrophication	55%	789%
Freshwater ecotoxicity	20%	3150%
Land transformation	3%	2496%
Water use	26%	20%
Resource use, minerals and metals	38%	347%
Resource use, fossil fuels	10%	28%
Total weighted results	25%	160%

sensitivity analysis, the 50/50 method instead of the cut-off approach is used. The 50/50 method assigns the credits and the burdens due to recycling to both previous (50%) and subsequent life cycle (50%). Accordingly, we allocate 50% of the impact of the primary production of vegetable oil to UCO. This method is the most conservative and it is usually applied for open-loop recycling when it is not known whether the use of the recycled material should be promoted (Schrijvers, Loubet, and Sonnemann, 2016).

Table 5 shows the variation of the environmental impact when the 50/50 method is applied to UCO. UCO is an oil waste derived from the use of oils and fats in cooking activities. For this sensitivity analysis, palm oil and soybean oil, whose data were available in the Ecoinvent database, were considered.[8]

When 50% of the environmental impacts of the production of the vegetable oils are allocated to the second life, the impacts of UCO-PP are significantly increased (see Table 6). On a weighted basis, the impacts of UCO-based PP would increase from 25% to 160% depending on the types of primary vegetable oil. In particular, larger variations are obtained when UCO origins from soybean oil. For climate change and the use of fossil resources, the environmental impact would increase by

17%−58% and 10–28%, respectively.

The second sensitivity is related to the multifunctionality of hydro-treatment and steam cracking. In the baseline calculations, energy allocation was used for hydrotreatment, while direct substitution for net steam and energy allocation is applied for the steam cracking process. Alternatively, different allocation methods could have been followed:

Energy allocation (only, not combined with direct substitution). In this approach, energy allocation is applied to all the co-products of steam cracking including steam. All the products have been valued with their lower heating values (LHVs) while the energy value of steam has been considered to be its enthalpy. Unlike the hybrid method applied for the steam cracking process in the baseline, no credits for steam substitution have been assigned (strictly attributional LCA and consistent with RED).

Exergy allocation (only, not combined with direct substitution). Compared to the baseline, the only difference is that exergy allocation has been applied to the steam cracking unit (for hydrotreatment using exergy or energy allocation key is indifferent). The reason behind this choice is that exergy can account for different quality in energy carriers. The superheated conditions of the steam released by steam cracking are 520 °C and 110 bars. This flow has been valorized with the exergetic value of this steam at such conditions. This means that it is assumed that this steam is entirely recovered and directly used, e.g. as process steam input in other refinery processes. The other co-products are energy and chemical products whose exergy value has been approximated

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[8] From Ecoinvent 3.4, Palm oil, crude {GLO}| market for | APOS, Soybean oil, crude {GLO}| market for | APOS.
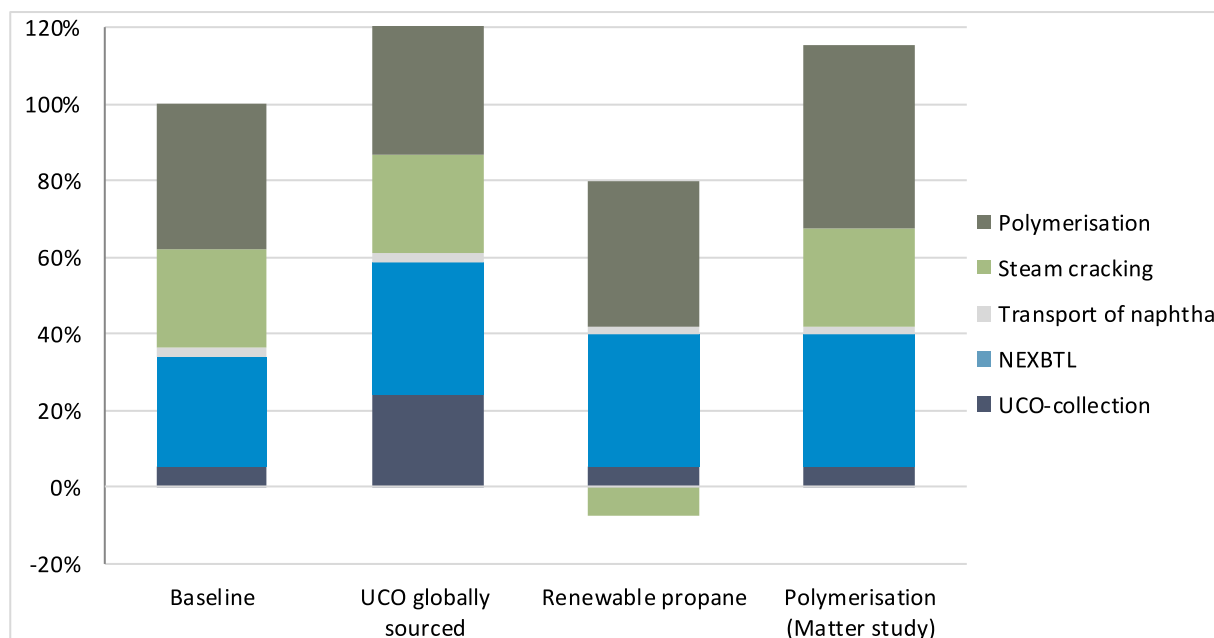
Fig. 3. Sensitivity analysis on data uncertainty (normalized and weighted results with baseline values taken as 100%). Steam cracking using renewable propane and methane is shown as a negative impact, as their production and combustion have a lower impact than the credit for steam production (substitution). For more details, see the main text.

with their LHVs. For the steam in input to the steam cracking unit, Ecoinvent database has been used[6].

Cut-off. Differently, from the baseline calculations, a cut-off approach is applied to the hydrotreatment unit process for bio-based naphtha. Due to the minor production share of bio-based naphtha compared to renewable diesel, all the environmental burdens of hydrotreatment are assigned to the renewable diesel. In this case, bio-based naphtha comes into the system as an 'emissions-free' input and, therefore, no impact has been apportioned for UCO collection and NEXBTL process. This is consistent with the model for this conversion route developed by the Joint Research center (JRC) of the European Commission (JRC, CONCAWE, EUCAR, 2014). In their model, HVO was the investigated product and the JRC applied a cut-off approach, neglecting the very small fraction of naphtha. The hybrid approach used for the steam cracking process remains unchanged.

Cut-off & energy allocation. In this case, the cut-off approach is applied to the hydrotreatment unit process. Differently from the previous case, solely energy allocation is applied to the steam cracking unit.

By-products substitution. In this case, system expansion followed by substitution is applied to all the by-products of hydrotreatment and steam cracking (but not to the co-products, i.e., diesel and ethylene). All the credits are assigned to the co-products diesel/naphtha and propylene/ethylene that is then partitioned by energy allocation. In the hydrotreatment unit, renewable propane has been assumed to replace petrochemical propane.[9] For the steam cracking unit, the following by-products are substituted with the conventional processes which would be avoided: steam, hydrogen, bio-methane, bio-based benzene, and bio-butadiene.[10] The other by-products (C5, C7, and C8) that have not been substituted have been considered as neutral, i.e., neither burdens or related credits are caused by them. For the baseline calculations, they

were instead accounted in the energy allocation shares.

Fig. 4 shows that energy and exergy allocation worsen the environmental footprint of bio-based PP. The only exception is resource use (minerals and metals) with energy allocation, although the difference is minimal (1%). This increase in impacts is caused by disregarding the credit from the substitution of steam in the baseline. Moreover, energy and exergy allocation lead to very similar results but impacts are higher in the case of exergy allocation. The reason behind this is that the exergy value of steam is lower than the enthalpy value. The weighted impact increases by 35%. Hence, it is concluded that exergy allocation is the most conservative among the assessed approaches.

The cut-off and by-product substitution approaches provide a significantly lower environmental footprint for bio-based PP. On the other hand, the by-products substitution approach is also the one assigning the highest impact for ionizing radiation HH and resource use of minerals and metals. The reason for this is that the credits for direct substitution of the by-products above do not compensate for the higher amount of impact apportioned to propylene. Moreover, negative impact results are obtained when system expansion followed by substitution is applied in the following impact categories: human toxicity without cancer effects, particulate matter, freshwater eutrophication, freshwater ecotoxicity and resource use of fossil fuels. This is caused by the by-products of hydrotreatment (propane) and steam cracking (especially bio-methane and benzene), which displace products that have high impacts for these five categories.

Conversely to other methods, the weighted impact decreases by (-) 113% by using the by-products substitution method (overall negative impact) compared do the baseline values. These negative flows violate the desirable characteristics of an attributional LCA (Majeau-Bettez et al., 2018). This negative impact means that the perturbation logic of substitution has created ''links between emissions and activities that are not mediated by product or service flows'' (Majeau-Bettez et al., 2018)

Moreover, the results presented in Fig. 4 confirm the findings of Sandin et al. (Sandin, Røyne, Berlin, Peters, and Svanström, 2015) that when a non-dominant product is in focus: (1) the results are sensitive to the choice of the allocation method and (2) the substitution method provides results in contrast with other allocation methods.

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[9] From Ecoinvent 3.4, Propane {GLO}| market for | Conseq
[10] From Ecoinvent 3.4, Steam, in chemical industry {RER}| production | Conseq, Natural gas, from medium pressure network (0.1-1 bar), at service station {GLO}| market for | Conseq. Butadiene {RER}| production | Conseq. From PlasticsEurope's Ecoprofiles, Hydrogen (reformer) E from PlasticsEurope (PlasticsEurope, 2005)), Benzene, at plant/RER based on PlasticsEurope Industry 2.0 database (PlasticsEurope, 2013)
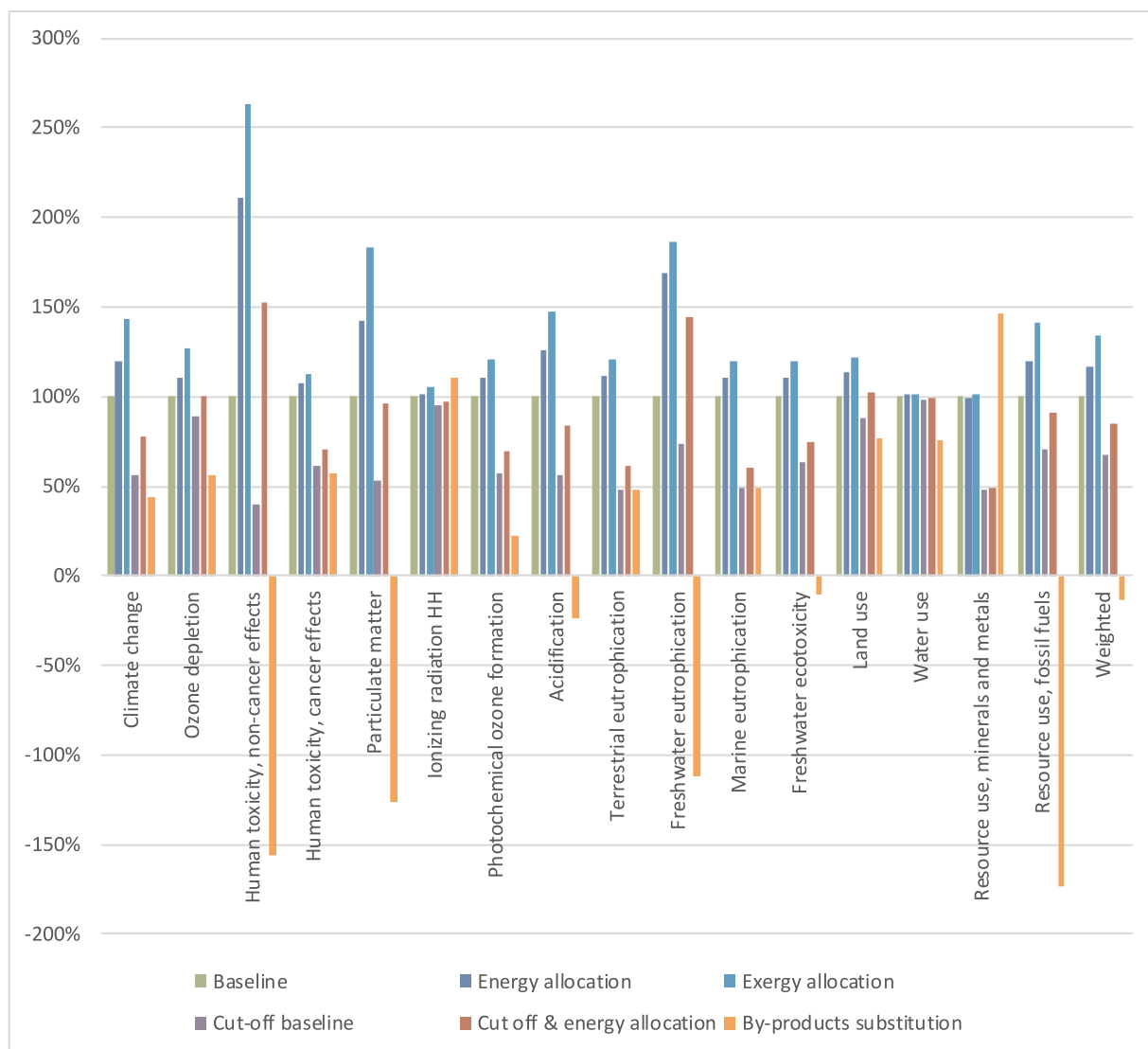
Fig. 4. Sensitivity analysis of the allocation approaches for hydrotreatment and steam cracking units (numerical values in Appendix B). Baseline results expressed as 100%.

These two findings apply only when non-dominant products are in focus. Thamsiriroj and Murphy, who studied the same route but with renewable diesel (HVO) in focus (the physically dominant product), concluded that UCO HVO is relatively unaffected by allocation methodology (energy allocation, substitution, and cut-off approaches) (Thamsiriroj and Murphy, 2011). Moreover, these results show that the RED statement that energy allocation provides results generally in line with the substitution method fails when a non-dominant product is investigated and cannot be extended to more impact categories than climate change.

The authors, therefore, recommend avoiding the use of the substitution method in attributional studies because its application can lead to results in contrast with other methods. The only exception can be the use of direct substitution for by-products when this can represent physical causality. Nevertheless, this should be based on a direct and empirically demonstrable relationship (Pelletier, Allacker, Pant, and Manfredi, 2014). This would also be in line with ISO 14044 recommending allocation by-physical causality shall be preferred to other allocation methods. When a mathematical model is not available, the use of direct substitution shall be validated by comparing it with other allocation (partitioning) methods. For this case study, the baseline results, where direct substitution has been applied, have been validated by comparison to energy, exergy, and cut-off allocation methods.

Moreover, the baseline results can be considered the most representative being an average among the possible allocation methods for all 16 impact categories and on a weighted basis.

4.3. Environmental benchmarking of UCO-based PP

The lack of harmonization in LCA method limits the direct comparison between bio-based and petrochemical materials studies, especially when considering multiple impact categories (Spierling et al., 2018). Specifically, such a comparison is often reliable only for climate change impacts (Spierling et al., 2018). In particular, this issue emerges when different black box datasets for petrochemical PP are compared (see Appendix C). From the analysis in Appendix C, it is possible to consider also resource use (fossil fuels) along with climate change as a reliable impact category (less than 10% variation). From cradle to factory gate, 1 kg of petrochemical PP causes 1.65–1.78 kg CO_2eq GHGs and the resource use (fossil fuels) ranges between 67 −74 MJ (see Appendix C). To be conservative, the two lower values have been used for comparison. Hence, UCO-based PP (baseline) has a 62% lower impact on climate change and 86% for resource use (fossil fuels) compared to petrochemical PP.

Moreover, these reductions are almost unaffected when UCO is globally imported (58% and 85% respectively). By using the 50/50

method, these reductions are reduced to the minimum of 40% and 82% respectively for climate change and fossil resources use when UCO is derived from soybean oil. When exergy allocation is used for hydro-treatment and steam cracking, these environmental impact reductions are instead reduced to a minimum of 45% in terms of climate change and to 80% as resource use of fossil fuels. From these results, it can be concluded that UCO-based PP is a favorable alternative option to pet-rochemical polypropylene in terms of climate change and fossil fuel resources.

It should be kept in mind that these impact reductions are for the cradle-to-factory gate scope and the differences above in climate change impact do not account for BCR. Thus, UCO-based PP has the potential advantages to act as a biogenic carbon sink if the material is recycled. Moreover, biogenic carbon emissions are released when burned in a waste-to-energy system. These advantages are out of the scope of this LCA and therefore not estimated.

Comparing our results with the LCA published by Kikuchi et al. (Kikuchi et al., 2017), it is found that UCO-based PP shows about 80/90% lower impact on climate change compared to bio-based PP made from sugarcane and woody biomass at factory-gate.

It should be recognised that UCO is a very limited feedstock. The European Commission already promotes its use for renewable diesel as a second-generation biofuel (European Commission, 2009). UCO-based PP is developed from the bio-based naphtha, which is a by-product of this renewable diesel. From an environmental perspective, it would be therefore interesting to assess what is more attractive between repla-cing petrochemical diesel or polypropylene. "Sidestream naphtha" case was chosen as baseline due to the current strong market demand for renewable diesel (main product). Nevertheless, the cracking of all HVO diesel and naphtha would be technically feasible. Increasing the mix-ture of bio-based HVO naphtha and diesel used for bio-based PP would not lead to a different environmental impact compared to the baseline calculation due to the slightly different LHVs assumed for bio-based naphtha and diesel (energy allocation). It is known that UCO renewable diesel could lead to GHG emissions saving up to 88%, which are much higher compared to 40–62% savings by other biodiesels (De Mora et al., 2015). These savings could appear higher than the ones allowed by UCO-based PP. Nevertheless, we are not able to answer this question properly without proper modeling for the end of life of polypropylene.

5. Conclusions

The first objective of the study is to identify the major environ-mental burdens in the cradle-to-factory gate life cycle of UCO-based PP. The environmental footprint of UCO-based PP is dominated by the polymerization process (38%), the production of hydrogen (21%), the production of LPG (18%) and the combustion (8%) of LPG. Climate change (28%), fossil resource use (23%) and water use (11%) have resulted in being the most important environmental impacts of UCO-based PP.

Compared to petrochemical PP, UCO-PP offers substantial impact savings for climate change (62%) and for fossil fuel resource use (86%). These savings remain substantial even in the cases of 1) globally im-ported UCO (58% and 85% respectively), 2) when UCO is considered a by-product instead of waste (40% and 82% respectively based on the 50/50 method) or 3) when a different allocation approach is used for hydrotreatment and steam cracking (savings of 45% and 80%, respec-tively). From these results, it can be concluded that bio-based PP from UCO is a promising alternative option to replace petrochemical poly-propylene in terms of climate change and fossil fuel resources.

It should be reminded that the comparisons made above are for the scope of cradle to factory gate. UCO-based PP has the further advantage of having a 100% biogenic carbon content embedded in the product, potentially for the long term (e.g. in a durable application). The full biogenic carbon balances should be accounted for in a future cradle to grave LCA when a final product made from UCO-PP is analysed.

The second objective of the study is to scrutinize how the allocation procedures used to solve multifunctionality affect the results of this LCA. It is found that exergy allocation leads to an increase of 35% of the environmental footprint of UCO-based PP compared to the baseline in which a hybrid direct substitution and energy allocation is applied. Conversely, the environmental footprint would become negative by using system expansion followed by substitution. The negative footprint obtained by using substitution is because bio-based naphtha and pro-pylene, which are the precursors of UCO-based PP, are two physically non-dominant output-products of multifunctional processes. Such a negative impact is a clear violation of the desirable characteristics of an attributional LCA. It is recommended to avoid the use of system ex-pansion followed by substitution in attributional studies. The only ex-ceptional cases are the ones where direct substitution of by-products can represent physical causality like applied to the steam produced during steam cracking. This steam produced is directly used by other processes of the same biorefinery, and, otherwise, should be produced as marginal production of refinery steam. In fact, this would result in line with ISO 14044 recommending allocation by physical causality shall be preferred to other allocation methods. As a mathematical model is not available to model physical causality relationships, the use of direct substitution for steam has been validated by comparing with other allocation methods, showing alignment in all the impact cate-gories assessed.

It is concluded that economic significance should be considered as an important requirement to fulfill before applying substitution. When this requirement is not respected, direct substitution shall be avoided.

Last but not least, UCO has been used as feedstock for a variety of applications of chemicals and fuels. The increasing demand for UCO has driven the price up in the past years. In this study, the impact of con-sidering UCO as a by-product instead of a waste was assessed. It is found that the LCA results could significantly vary depending on the type of original vegetable oils as well as how the allocation is performed (e.g. based on the 50/50 approach). The results provided in this study should be used to elicit the discussion in the context of assessing the impacts of products in a future circular and bio-based economy.

CRediT authorship contribution statement

Christian Moretti: Conceptualization, Methodology, Software, Data curation, Formal analysis, Investigation, Visualization, Writing - original draft, Writing - review & editing. **Martin Junginger:** Conceptualization, Data curation, Supervision, Funding acquisition, Resources, Writing - review & editing. **Li Shen:** Conceptualization, Methodology, Project administration, Funding acquisition, Resources, Supervision, Writing - original draft, Writing - review & editing.

Declaration of Competing Interest

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

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2020.104750.

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