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Green Chemicals from Used Cooking Oils: Trends, Challenges and Opportunities

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ABSTRACT

Food waste reduction is fundamental for sustainable development and pursuing this goal, recycling and the valorization of used cooking oil (UCO) can play a major contribution. Although it has been traditionally used for biofuels production, the oleochemical potential of UCOs is vast. UCOs can be used as feedstock for a large variety of value added green chemicals including plasticizers, binders, epoxides, surfactants, lubricants, polymers, biomaterials, and different building blocks. Thus, UCOs transformation into functional chemicals can bring long-term stability to the supply chain, avoiding the current dependence on commodity products. In this regard, this work describes some of the potential benefits of using UCOs as feedstock in oleochemical biorefineries. Also, some of the most recent investigations on the valorization of UCOs other than biofuel are presented. Finally, major challenges and future directions are discussed.

Keywords. Used cooking oil, biobased chemicals, value-added products, challenges

1. Introduction

Reduction of food loss and waste is paramount to fulfil the UN's Sustainable Development Goals, and crucial to curtail their associated economic, social and environmental life cycle impacts. Current estimations indicate that average per capita food waste generation in Europe ranges between 173 - 290 kg/person·yr [1]. Equally alarming numbers (in kg/person·yr) are observed in Australia (361), USA (278), Canada (123), India (51), China (44), and other countries [2]. Reported data also reveals that nearly 60% of food waste is generated as consumption and post-consumption residues (e.g. bones, cooking oils, peels, leftovers, etc.), and a large fraction is unavoidable or inedible [1]. In order to mitigate the impacts, different circular economy approaches have been proposed for the exploitation and valorization of food waste via transformation into a large variety of chemicals, materials and fuels through a biorefinery

35 approach [3-6]. The potential valorization processes and the targeted products largely depend on the
36 nature and composition of waste. Besides carbohydrates, starches, and proteins, a large fraction of typical
37 food residues corresponds to fats and oils (i.e. up to 25% wt. on a dry basis, [4, 7]). Particularly, among
38 the different food wastes, discarded used cooking oil (UCO) is a major source of lipids. While suitable
39 processes are required to extract the lipid content from most food waste [7], lipids in UCOs are readily
40 reachable. This explains the existing UCOs collection chains and the different processes for their
41 valorization at an industrial scale [8-10].

42

43 UCOs are mainly generated in households and hospitality sectors (HORECA – Hotels, Restaurants,
44 Casino-Cafe-Catering) [11, 12], and the current global production is estimated between 20 and 32% of
45 total vegetable oil consumption (41 - 52 Mt/yr., [9, 13]). This broad range is a result of the different
46 culinary customs and consumption trends in the different regions, which also play a major role in the
47 nature, chemical composition, and content of impurities in the UCOs [8]. Because of unconscious
48 behaviors, absence of regulations, or lack of law enforcement, most UCOs are generally disposed through
49 sinks and syphons, or within the solid residues that are sent to landfills. In addition to ecosystems
50 pollution and public health impacts, this mismanagement generates a variety of cascading problems
51 including sewage clogs, wastewater overflow, costly damage to infrastructure, vectors and pests,
52 nauseous odors, higher operating costs at central wastewater treatment plants, etc. In order to mitigate all
53 these problems, a fraction of UCOs have been typically recovered and reused as oleochemical feedstock,
54 mainly for the production of low added-value commodities such as biofuels (e.g. biodiesel, hydrogenated
55 vegetable oil - HVO), soaps and animal feed. This has created a small but solid market of nearly 600
56 Million USD/yr., growing at an average annual rate of 4% [14], and with prices in the range from 620-
57 865 USD/t during the last 12 months [15]. In spite of the growing trend, and because of the low added-
58 value of current derivatives, UCOs market is highly vulnerable to the change of economic and political
59 environment. This vulnerability could be reduced by diversifying the portfolio including high value-added
60 byproducts, thus bringing long-term stability into the entire valorization chain. In this regard, this work
61 describes most current research on the transformation of UCOs into high value added biobased products,
62 the challenges for a successful industrial implementation, the associated benefits, and some future
63 directions.

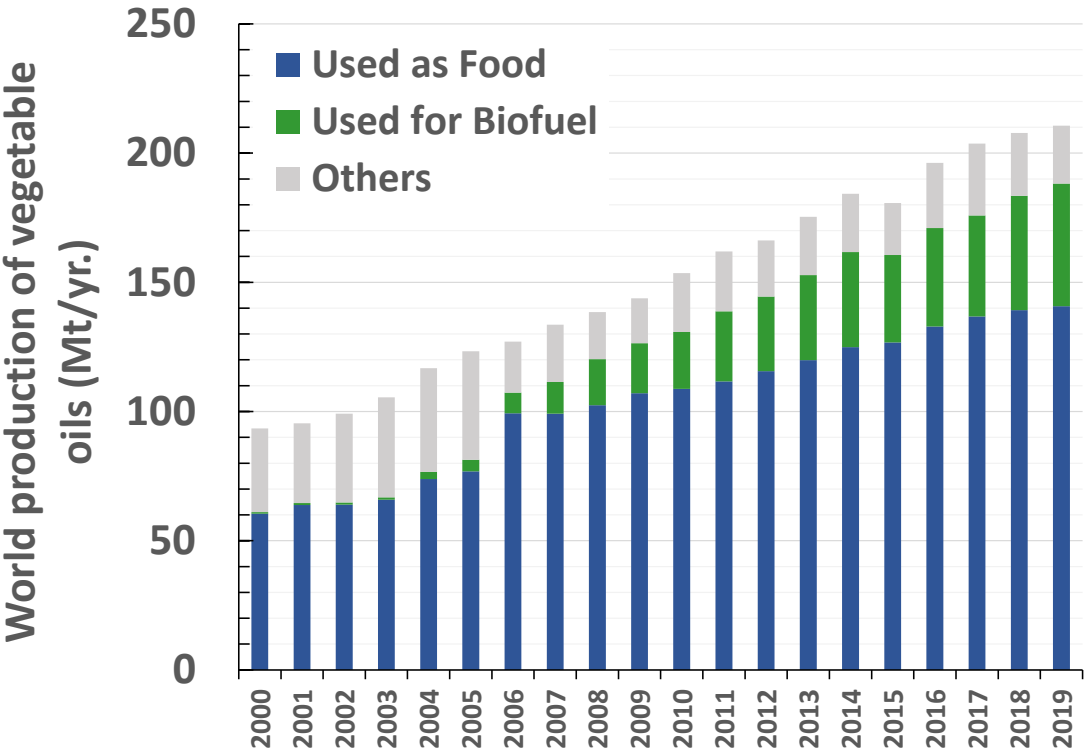
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65 **2. Potential benefits of UCOs as Oleochemical feedstock**

66

67 The main raw materials of the oleochemical industry are vegetable oils and animal fats, with the former
68 having a 99.9% share in volume, and the remaining small fraction corresponding to butter, fish oils, and

69 fats from animal rendering [16]. Figure 1 presents the historical production of vegetable oils and the
 70 corresponding distribution regarding final use. As observed, 68% of current world production is used for
 71 food applications (i.e. cooking oils, food ingredients), and 23% in biofuels, mainly biodiesel
 72 (~1.1kg_{Oil}/1kg_{Biodiesel}). The remaining fraction (~9%) is destined for feed and other oleochemical uses
 73 including drop-in applications (e.g. additive for polymers, resins, asphalt, lubricants, greases, drying oils,
 74 rubber products, etc.) and as feedstock for different chemical derivatives. Taking into account the
 75 estimated global UCO generation (41 Mt, [9]), this amount can replace the virgin vegetable oil currently
 76 required as feedstock for the oleochemical industry, a part of which is used as a biodiesel feedstock.
 77 Hence, the exploitation of UCOs as chemical feedstock within a circular economy model would help to
 78 reduce the environmental and social impacts associated to both, the edible oil and the oleochemical
 79 industries.

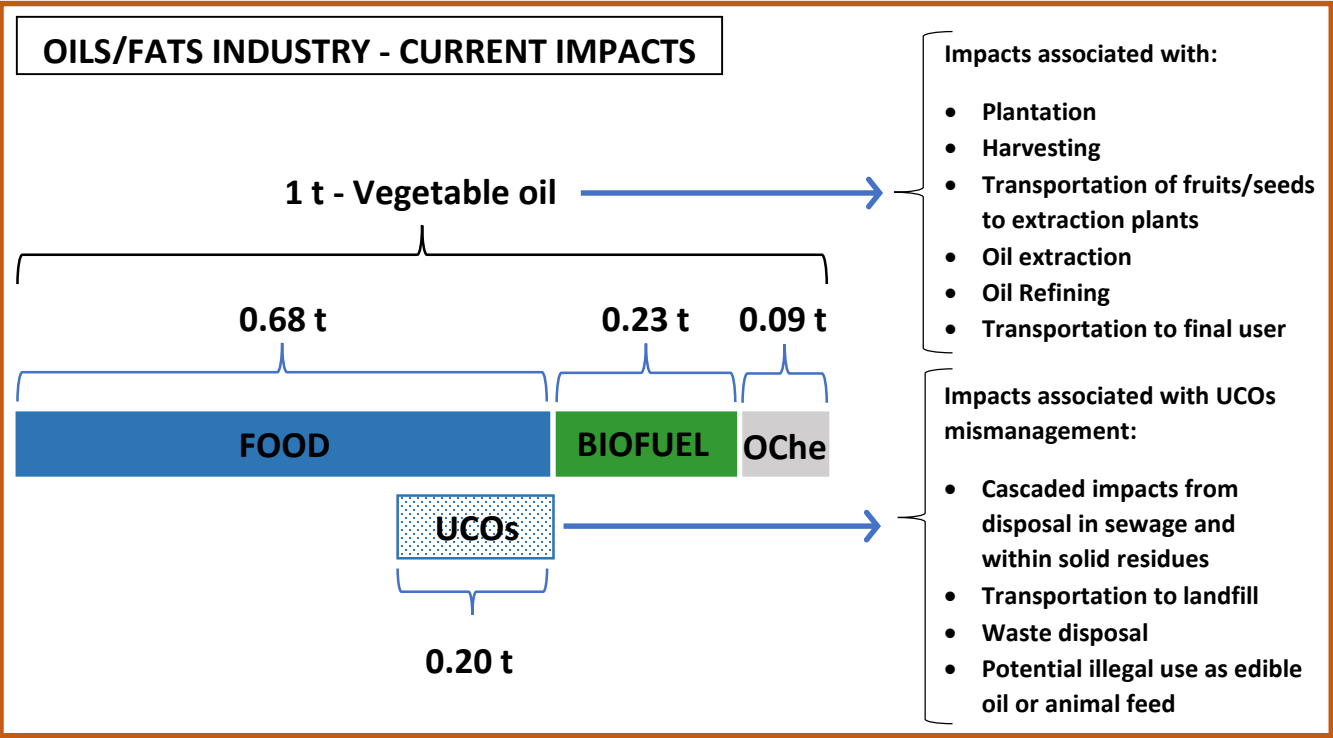


98 **Figure 1.** Historical world production of vegetable oils and share among final uses (Data from [16]).

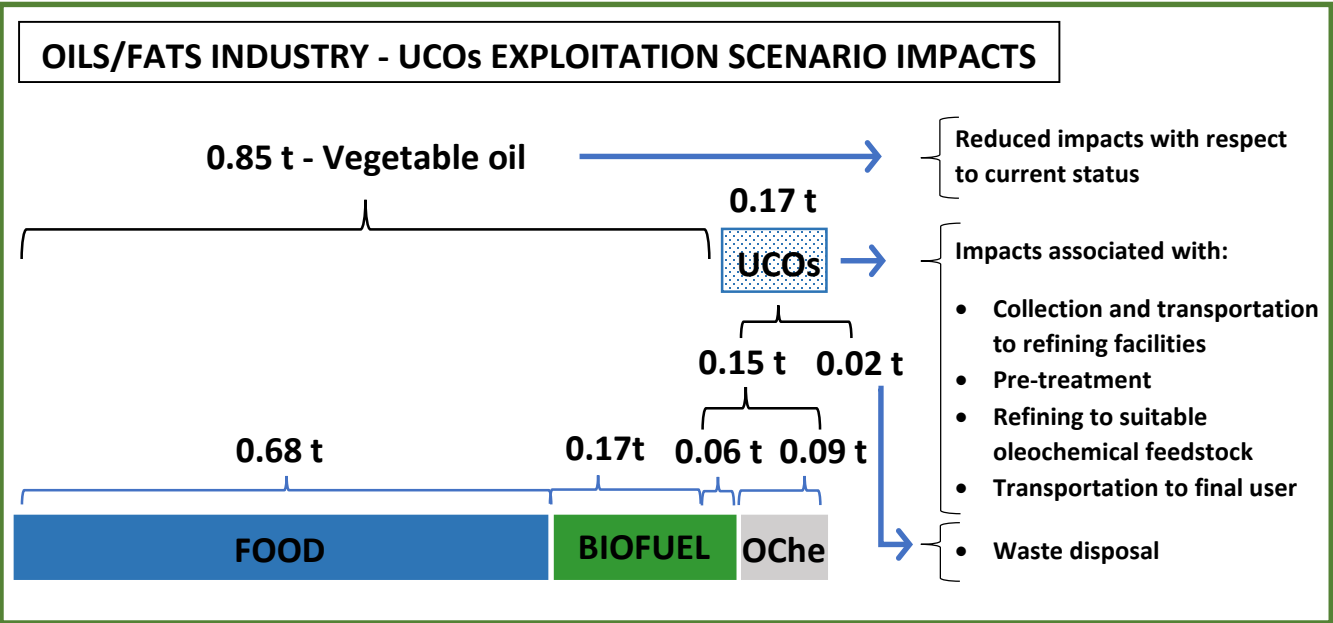
100 Figure 2 schematically presents the different stages in which negative social and environmental lifecycle
 101 impacts could be mitigated by using UCOs as oleochemical feedstock [17-22]. This is based on a 100%
 102 efficiency in UCO collection and reuse, and considering the more conservative estimation of UCOs
 103 generation with respect to vegetable oil consumption (20%, [9]). Also, it is important to consider that for

104 instance in the EU, only 45-50% of collectable UCO is recovered, with a best-case scenario of 70% [11,
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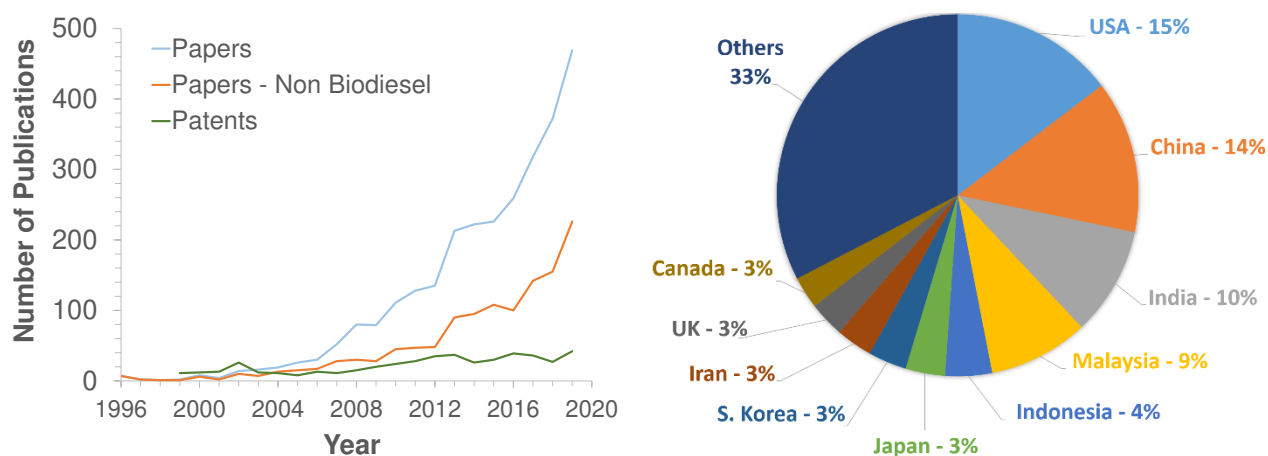
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136 **Figure 2.** Potential impacts mitigation of employing a circular economy approach in the exploitation of
 137 UCOs as oleochemical (OChe) feedstock considering lifecycle stages (Cradle-to-gate.). Functional unit: 1
 138 t vegetable oil. Assuming a 90% yield of refined UCO from the collected one.
 139

140 3. High value added application for UCOs

141
142 The valorization of UCOs via transformation into suitable oleochemical products have captured the
143 attention from academic and industrial researchers during the last two decades. Figure 3 presents the
144 evolution of the scientific production (i.e. papers and patents) dealing with the exploitation of UCOs. While
145 the studies on biodiesel are still predominant, there is an increasing trend to explore novel applications,
146 mainly focused on value-added products. In addition to the availability of financial resources, most research
147 have been conducted in countries where UCOs mismanagement can be a major problem, either because of
148 their large population (e.g. China, India), or for the large per capita generation (e.g. USA, Indonesia, S.
149 Korea). Most EU countries are grouped as “others”, and in this case, their large scientific productivity has
150 been promoted by the public policies of the community [20].



161
162 **Figure 3.** Number of publications and patents on the valorization of used cooking oil in last decades, and
163 share by major contributing countries (March, 2020). Searching terms: TITLE-ABS-KEY ("Used
164 cooking oil" OR "Waste cooking oil" OR "Yellow grease" OR "Brown grease" OR "Trap grease")
165 (Source: [24-26]).
166

167 Recent reports indicate that biobased products can have a large market growth in the coming years, if similar
168 policies and subventions to those implemented to the production of biofuels, are also implemented for green
169 chemicals [27]. In this context, a variety of new processes and products have been developed for UCOs
170 valorization, evolving from basic drop-in applications, to more complex thermochemical, chemical and
171 biochemical transformations [28-31]. More recently, further exploration has enabled the development of
172 value added products from the crude glycerol obtained as co-product from UCOs-based biodiesel processes
173 [32, 33]. While this is not intended to be a comprehensive review of the available literature, Table 1 presents
174 a summary of the most recent attempts for UCOs harnessing, including the production of plasticizers,
175 binders, epoxides, surfactants, lubricants, polymers, biomaterials, building blocks etc.

Table 1. Most current attempts on the production of biobased chemicals from UCOs and UCOs-based glycerol

Application	Process	Product	Chemistry behind product use	Highlights	Ref.
UCOs Valorization					
Plasticizer	Transesterification of UCOs biodiesel with 2-ethylhexanol and further epoxidation	Epoxidized 2-ethylhexyl fatty ester	Oxirane reacts with compounds containing active hydrogen atoms (e.g. water, organic acids, alcohols, Halides)	Ep-WCOEtHEs in PVC enhanced the overall mechanical property and thermal stability, with no significant change in migration-resistant performance.	[34]
	Epoxidation	Epoxidized UCO	Enhanced thermo-oxidative stability by reducing unsaturations	Primary plasticizer for PVC films, without the need of other additives, resulting in samples with good thermal stability and mechanical properties	[35]
	Transesterification of UCO with methanol and epoxidation of methyl ester. Esterification with citric acid, and final acetylation with acetic anhydride	Acetylated FAME citric acid ester (Ac-FAME-CAE)	Hydrogen bonding of the 8 carboxylic groups of Ac-FAME-CAE with PVC polymer to enhance thermal stability	Similar plasticizing performance to DOP	[36]
	Epoxidation	Epoxidized UCO	Oxirane reacts with compounds containing active hydrogen atoms (e.g. water, organic acids, alcohols, Halides)	Homogeneous and heterogeneous catalysts were tested. Dimers and oligomers formed using H ₂ SO ₄ as catalyst	[37]
	Esterification and transesterification with methanol and amino-methylation (Mannich reaction)	Mannich base of UCO biodiesel	chlorine atoms of PVC substituted with Mannich base of UCO biodiesel	Lower thermal stability due to the content of active secondary amine group	[38]
Asphalt/pavement binder	Drop-in. 5%wt. addition	Asphalt binder with light components from UCO	Carbonyl groups reacting with binders	Low temperature crack resistance and softness of the asphalt binder are improved	[39]
	Drop-in. 0.4 - 0.8 %wt. addition	Macadam pavement	UCO physically cover aggregates	Improves cracking and fatigue resistance	[40]
	Co-pyrolysis of UCO with rubber	Rubber/UCO binder	UCO reacts with rubber polymers during de-sulfurization and pyrolysis	Improved low temperature properties of binder Improved rheological properties of asphalt	[41, 42]
	Drop-in. 5%wt. addition	Binder replacement	Unsaturations bond with asphalt macromolecules and binder	Treated waste cooking oil can be used as a replacement of asphalt binder in asphalt mixtures	[43, 44]
	Drop-in. 5% UCO addition	Asphalt binder	Rheological modifier of asphalt binder	Addition of waste cooking oil as binder replacement was improve the durability performance of asphalt mixture.	[45]
Masonry binder	Drop-in. 10%wt. addition	Construction block	Oxy-polymerization and crosslinking	WasteVege block does not require the use of any form of cementitious or pozzolanic materials or water.	[46]

Epoxidized biodiesel	Enzymatic transesterification and epoxidation	Epoxidized UCO biodiesel	Oxirane reacts with compounds containing active hydrogen atoms (e.g. water, organic acids, alcohols, Halides)	Impurities had a negative effect in the epoxidation. Oxirane value 2.5.	[47]
Polyol / Polyurethane	Epoxidation of UCO and hydroxylation with diethylene glycol	UCO-based polyol	Hydroxyl groups react with isocyanate to form urethane bonds	The application of sulfuric acid in this experiment required a much higher temperature than in the case of the catalysts based on tetrafluoroboric acid and a longer reaction time. Satisfactory polyurethane bio-foams can be produced by replacing 40-60% of polyol with biobased alternative	[48, 49, 50]
Lubricant	Epoxidation and hydroxylation with methanol, ethanol, and 2-ethyl hexanol, esterification with hexanoic anhydride	UCO and UCO FAME poliol hexanoic ester	Enhanced thermo-oxidative stability by reducing unsaturations and hydroxyl groups	Products are compliant to standard lubricant specifications in terms of viscosity, viscosity index and pour point; with much higher biodegradability	[51]
	Epoxidation of UCO	Epoxidized UCO	Enhanced thermo-oxidative stability by reducing unsaturations	Epoxidized UCO exhibit highly desirable and enhanced physicochemical properties in all the aspects for environmentally friendly biolubricants.	[52]
	Enzymatic hydrolysis and esterification	Fatty acid neopentyl glycol ester	Enhance lubricity taking into account viscosity profile	A maximum conversion of 94% was found after 24 h using immobilized enzyme	[53]
	Drop-in. 15%wt. addition	UCO dispersible Cu nanoparticles	Unsaturations complexed on the nanoparticles surface.	Particles are synthesized in UCO, and directly used as additive. The formulations are stable, without segregation even after months.	[54]
Surfactant	Transesterification with methanol, sulfonation of methyl ester, and neutralization with NaOH	Methyl ester sodium sulfonate	Sulfonation provides polar moieties to FAME turning it into surfactant	Yield of methyl ester sulfonic acid (MESA) after sulfonation was obtained 77.20%. Methanol reduced substitution of methyl groups into a disalt	[55]
	Saponification of UCO with KOH, Acidification to FA, esterification with methanol to FAME, reduction to Fatty alcohol, esterification with chloroacetic acid, and amination	Diaminium chloride gemini-surfactant	Negatively charged carbonylic oxygen and chloride from surfactant are adsorbed on metal surface creating a protecting barrier layer	Efficacious inhibitor for steel (N80) corrosion	[56]
Liquid detergent	Transesterification with methanol and sulfonation of methyl ester	Methyl ester sulfonate	Sulfonation provides polar moieties to FAME turning it into surfactant	Liquid detergent comprising of 15% MES concentration and 0.1% ZnO nanoparticles	[57]
Biopolymer precursors	Transesterification and ethenolysis	Ethenolyzed and self-metathesized products	Olefinic bonds in reaction products can be used for further polymerization	A novel renewable lipidic source of spent hen for ethenolysis is exploited for the first time	[58]

Biobased Polymers	Epoxidation, hydroxylation with water, polymerization with Methylene diphenyl diisocyanate	UCO-based polyurethane doped with lithium iodide	Hydroxyl groups react with isocyanate to form urethane bonds	UCO-based PU could be used as a potential host for polymer electrolyte	[59]
	Fermentation	Polyhydroxyalkanoate and astaxanthin-rich carotenoids	Biodegradable, elastomeric, thermoplastic and biocompatible polymer	1% v/v UCO was used. 1 g/L of PHA	[60]
	Fermentation	Polyhydroxybutyrate [P(3HB)]		WCO could provide better accumulation of P(3HB) in <i>C. necator</i> H16 compared to other common plant-based oil. The higher production of P(3HB) was approximately 80 wt%.	[61]
	Fermentation	polyhydroxyalkanoates (PHAs) - (R)-3 hydroxyoctanoic acid and (R)-3-hydroxydecanoic acid monomers		Low molecular weight 18342 kDa, Low yields probably caused by inhibitory compounds in UCO	[62]
Fermentation supplement	Drop-in. 2-3%wt. addition	Microbial oil		Carbon source for biomass	Mixture of UCO with crude glycerol enhances oil accumulation in yeast
	Fermentation	Lipase	maximum lipase activity (12 000U/L), also lipid-rich biomass (48% of lipids mass per dry cellular mass),		[64]
	Fermentation	d- and l-Limonene	UCO is superior substrate than glucose, but low titers obtained 2.5-2.7 mg/L		[65]
Structured materials	Double thermal chemical vapor deposition	Graphene	Pyrolysed UCO is a carbon source for graphene formation	Utilization of UCO as the carbon source for the formation of multilayer graphene has been successfully performed.	[66]
3D printing resin	Acrylation	Triacylglycerol acrylate	UV-promoted crosslinking of acrylic moieties in acrylated UCO	Higher biodegradability of printed plastics, no photo inhibitors required	[67]
Emulsion liquid membrane	Drop-in. 50-80%wt. addition	Emulsion	UCO is used as organic solvent of liquid emulsion	99.1 efficiency in the recovery of organic dyes from contaminated water	[68]
Flotation oil	Pyrolysis	Deoxygenated hydrocarbons	Adsorption of hydrocarbons on coal surface	UCO pyrolysis products possessed strong collecting ability and better selectivity, and can replace diesel as a coal flotation collector.	[69]
Bioadsorbent	Impregnation and pyrolysis	Ordered micro-mesoporous carbon nanocasted on HZSM-5/SBA-15	Carbonaceous material contains oxygen-rich groups suitable for adsorption of cationic dyes. Ordered micro-mesoporous structure combines size selectivity and high diffusion rates	Material exhibits high adsorption capacity comparable to activated carbons	[70]

UCOs-based Glycerol valorization					
Carbon source	Fermentation	Hydrogen	Biobased building blocks	Bioconversion of crude glycerol by sub-tropical mixed and pure cultures. 15.14 mol H ₂ /mol glycerol	[71]
		Lactic acid		Bioconversion to lactic acid by <i>Rhizopus microsporus</i> . Lactic acid average production of 3.99 g/L	[72]
		1,3 Propanediol		The effect of crude glycerol impurities on 1,3-propanediol biosynthesis by <i>Klebsiella pneumoniae</i> DSMZ 2026. 9.69 g/L 1,3-PD (yield 0.21 g/g, productivity 0.80 g/L/h) was obtained after 12 h	[73]
		1,3 Propanediol		Production to 1,3-Propanediol and Lactate by a Microbial Consortium. Impurities in GWCO did pose a great challenge to microbial growth and metabolism. In fed batch fermentation, 27.77 g/L 1,3-PDO and 14.68 g/L LA were achieved.	[74]
		Valeric acid		Anaerobic fermentation with open microbiome. High valerate extraction rates with medium and maximum values of 12.9 and 30.0 g COD /m ³ day, were obtained with low ethanol addition (15% of the glycerol-COD)	[75]
		1,3 Propanediol		1,3-PDO production with a mixed culture, A maximum productivity of 7.49 g/Ld	[76]
		Lipids		Lipid production via fermentation with <i>Trichosporon oleaginosus</i> . The highest lipid yield 0.19 g/g glycerol was obtained at 50 g/L purified glycerol in which the biomass concentration and lipid content were 10.75 g/L and 47% w/w, respectively.	[77]
		Citric acid, Succinic acid		A Suitable Substrate for the Growth of <i>Candida zeylanoides</i> Yeast Strain ATCC 20367. Biosynthesis of organic acids (e.g., citric 0.66 g/L; and succinic, 0.6 g/L) was significantly lower compared to pure glycerol and glucose used as main carbon sources.	[78]

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178

179 4. Challenges and future directions

180
181 As observed, UCOs can be used as raw material for a large variety of green chemicals. In addition to the
182 technical limitations observed in some of the processes, there are a number of issues that need to be
183 overcome in order to enable industrial implementation. As in any other biorefinery, the supply chain plays
184 a major role in the sustainability of the proposed production schemes. In this case, a major fraction of the
185 generated UCOs comes from the Household segment, for which very low recovery efficiencies are typical
186 (< 6%, [11]). Hence, it is necessary to deploy effective policies and regulated practices to enhance UCOs
187 recycling and collection rates, under multi-stakeholders considerations (i.e. authorities, generators,
188 collecting companies, biorefineries). This also indicates that there is need to optimize the collection
189 schemes to ensure that the consumed resources (e.g. energy, financial) do not surpass those from the
190 obtained UCOs, mainly when the biorefineries operate as centralized facilities far from the source. For
191 instance, one study has shown that from a life cycle perspective, biodiesel from European rapeseed UCOs
192 is less sustainable than petroleum diesel and even less than biodiesel from Indonesian's palm oil UCOs
193 [18].

194
195 Another major challenge is the highly heterogeneous nature of UCOs. They exhibit a large variability in
196 physicochemical and sensory properties, and a substantial amount of impurities [8, 79, 80], resulting from
197 different diets, culinary practices, and management procedures. In most of the studied processes of Table 1
198 there were reports of impurities in the raw material affecting the catalytic and biologically conversion steps,
199 the drop-in uses, and even the thermochemical transformations. Also, unpleasant sensory properties (e.g.
200 color, appearance, odour) are of major concern. Therefore, suitable upgrading processes must be
201 implemented to enable the efficient transformation of UCOs to the desired biobased chemicals without
202 compromising the economic feasibility [81]. Also, resilient and intensified technologies must be developed
203 to enable the use of other types of waste lipids (e.g. trap grease, rancid oils, food/solid waste lipids, etc.).
204 In any case, even after pretreatment and upgrading, the presence of trace impurities also might prevent that
205 some of the derivatives could be used in sensitive applications (e.g. personal care products, cosmetics, food
206 or pharmaceuticals) where the market is more attractive. Alternatively, they could be directed to other
207 markets such as construction materials, asphalt, rubbers, lubricants, surfactants, fuels, etc.

208
209 A current threat to the industrial implementation of UCOs-based chemicals, is that they are strongly linked
210 to the biodiesel market, and there are some concerns about the sustainability of this fuel especially given
211 the rapid move from liquid fuel to electric-powered vehicles. Nowadays, UCOs biodiesel is promoted via
212 public policies such as the Renewable Energy Directive (RED II) from EU. According to this directive,

213 UCO biodiesel can be double-counted, so its price can be higher than first generation biodiesel, encouraging
214 supply and demand. Nevertheless, recent claims indicate that at least one-third of UCO-based biodiesel in
215 the European market is fraudulent, because apparently it corresponds to the recently banned palm oil
216 biodiesel [82]. Besides, some of the unsustainable palm oil that was prohibited in the EU has been diverted
217 to China for animal feed in order to replace the UCOs that are currently exported to Europe [83]. These
218 type of problems might push for revisions of RED II, which will directly affect UCOs supply for the
219 oleochemical industry [84]. Finally, current COVID-19 pandemic is putting pressure on UCOs global
220 trading, affecting supply, dropping prices, and reducing the potential profitability of the biorefineries [85].

221

222 **5. Concluding Remarks**

223

224 Used Cooking Oil is a valuable food waste that can be transformed into a large variety of products. While
225 the use as biofuel feedstock enabled the creation of a global collection and supply chain of UCOs, only the
226 incorporation of high value added green chemicals within the biorefineries would ensure their long-term
227 sustainability. As presented, UCOs exploitation as oleochemical feedstock can involve large reductions in
228 life cycle impacts, cutting the need for virgin vegetable oil, and alleviating the impacts of the current
229 mismanaging practices for disposal. Also, by using UCO derivatives as ingredients in different end
230 products, there is a contribution to “green” other sectors such as polymers, asphalts, cementing materials,
231 detergents, lubricants, etc. Despite such a circular economy model around UCO seeming attractive, major
232 challenges have to be overcome. Future developments will be mostly focused on dealing with UCOs
233 heterogeneity and impurities content, upgrading processes, enhancing household collection, and
234 implementing resilient and intensified processes capable of incorporating different types of waste lipids
235 (e.g. trap grease, rancid oils, solid waste lipids, etc.).

236

237 **Conflict of interest statement**

238 Nothing declared.

239

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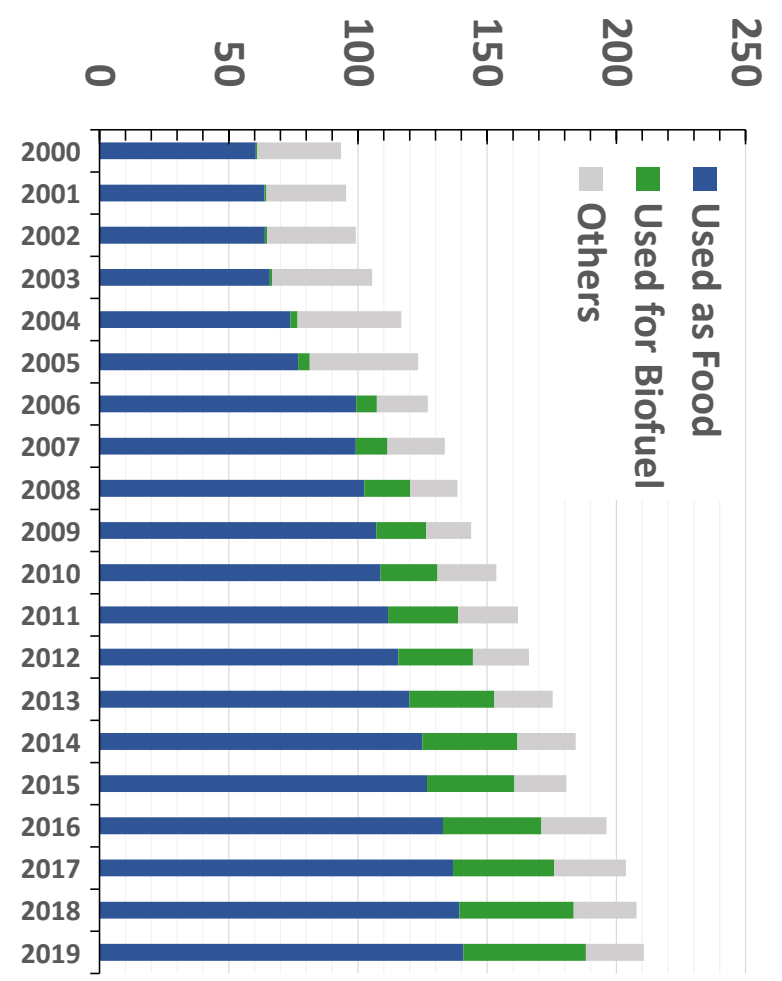
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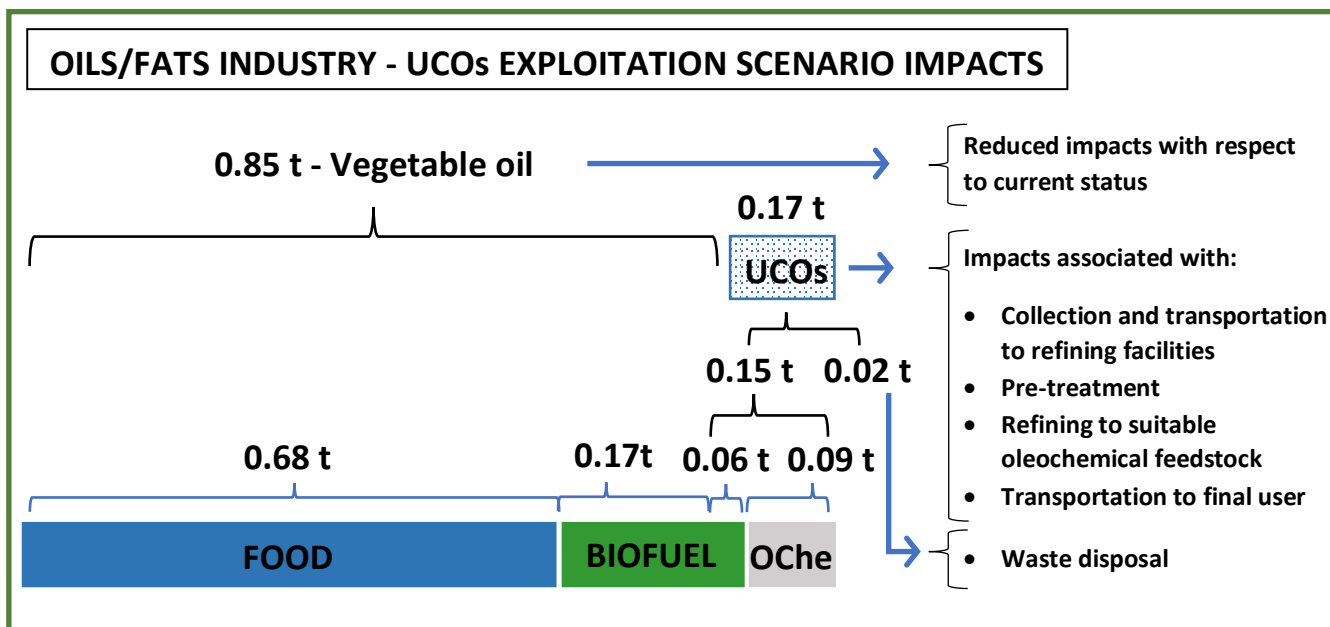
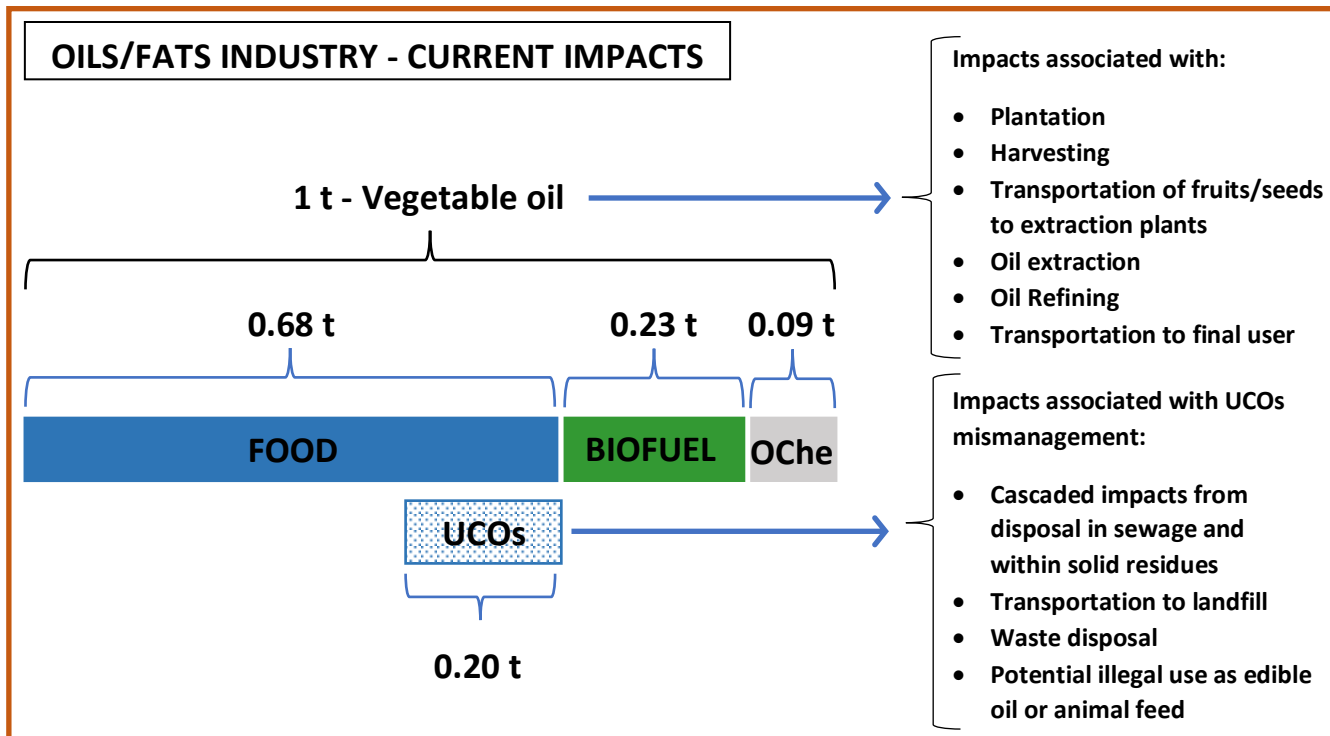
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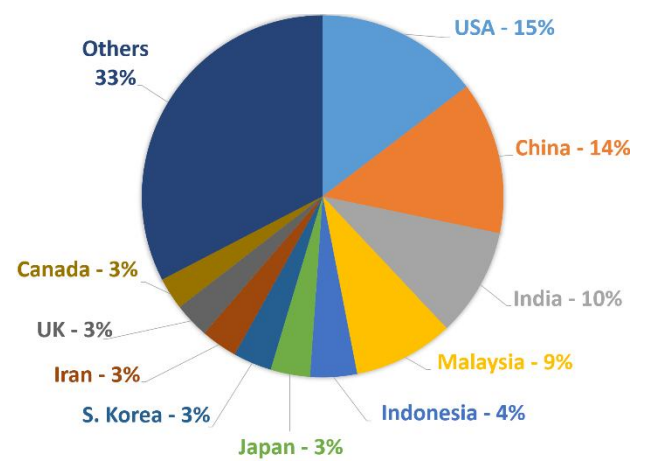
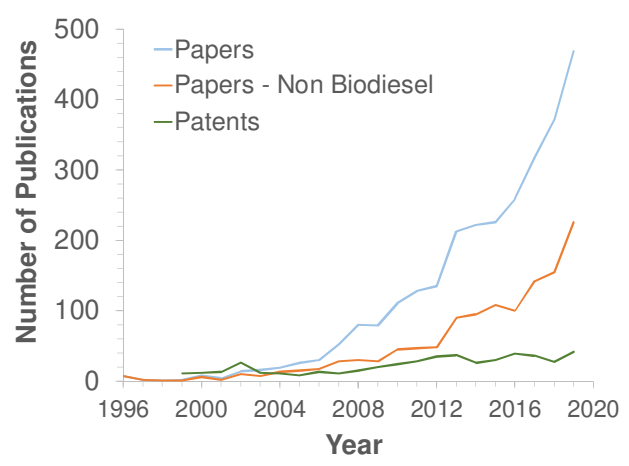
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World production of vegetable oils (Mt/yr.)





Figure



HIGHLIGHTS

- World production of used cooking oils (UCOs) and current market data
- Identification of life cycle impacts reduction by exploitation of UCOs
- Review of current state of art on used cooking oil valorisation into green chemicals
- Major challenges on UCOs utilization as oleochemical feedstock