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Current Opinion in
Green and Sustainable Chemistry

Green chemicals from used cooking oils: Trends, challenges, and opportunities

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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 biofuel 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, UCO 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. In addition, some of the most recent investigations on the valorization of UCOs other than biofuel are presented. Finally, major challenges and future directions are discussed.

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Current Opinion in Green and Sustainable Chemistry 2020, **26:**100369

This review comes from a themed issue on **Special Issue on South America**

Edited by **Vania Zuin and Rodrigo O.M.A. de Souza**

Available online 7 June 2020

For complete overview of the section, please refer the article collection - Special Issue on South America

https://doi.org/10.1016/j.cogsc.2020.100369

Introduction

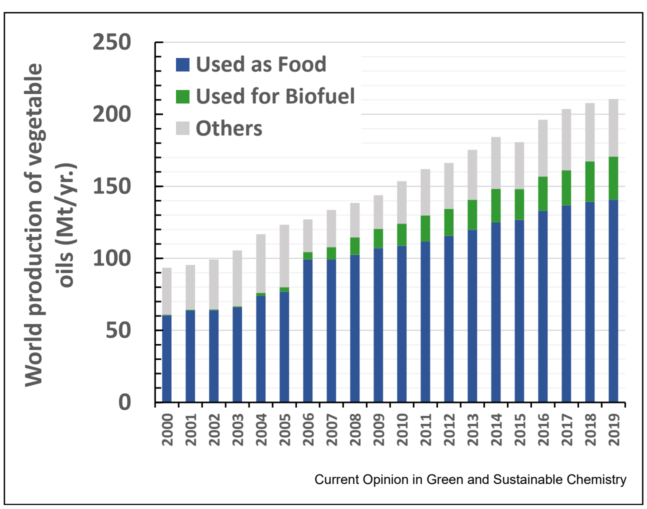
Reduction of food loss and waste is paramount to fulfill the UN's Sustainable Development Goals and crucial to curtail their associated economic, social, and environmental life cycle impacts. Current estimations indicate that average per capita food waste generation in Europe ranges between 173 and 290 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 reveal that nearly 60% of food waste is generated as consumption and postconsumption residues (e.g. bones, cooking oils, peels, leftovers, and so on), and a large fraction is unavoidable or inedible [1]. 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 approach [3–6]. The potential valorization processes and the targeted products largely depend on the nature and composition of waste. Besides carbohydrates, starches, and proteins, a large fraction of typical food residues corresponds to fats and oils (i.e. up to 25% wt. on a dry basis [4,7]). Particularly, among the different food wastes, discarded used cooking oil (UCO) is a major source of lipids. While suitable processes are required to extract the lipid content from most food waste [7], lipids in UCOs are readily reachable. This explains the existing UCO collection chains and the different processes for their valorization at an industrial scale [8–10].

UCOs are mainly generated in households and hospitality sectors (hotels, restaurants, casinos, cafes, catering) [11,12], and the current global production is estimated between 20% and 32% of total vegetable oil consumption (41–67 Mt/yr [9,13]). This broad range is a result of the different culinary customs and consumption trends in the different regions, which also play a major role in the nature, chemical composition, and content of impurities in the UCOs [8]. Because of unconscious behaviors, absence of regulations, or lack of law enforcement, most UCOs are generally disposed through sinks and syphons, or within the solid residues that are sent to landfills. In addition to ecosystem pollution and public health impacts, this mismanagement generates a variety of cascading problems including sewage clogs, wastewater overflow, costly damage to infrastructure, vectors and pests, nauseous odors, higher operating costs at central wastewater treatment plants, and so on. To mitigate all these problems, a fraction of UCOs have been typically recovered and reused as oleochemical feedstock, mainly for the production of low added-value commodities such as biofuels (e.g. biodiesel, hydrogenated vegetable oil), soaps, and animal feed. This has created a small but solid market of nearly 600 million USD/yr., growing at an average annual rate of 4% [14], and with prices in the range from 620 to 865 USD/t during the last 12 months [15]. Despite the growing trend and because of the low added value of current derivatives, UCO market is highly vulnerable to the change of economic and political environment. This vulnerability could be reduced by diversifying the portfolio including high value-added byproducts, thus bringing long-term stability into the entire valorization chain. In this regard, this work describes most current research on the transformation of UCOs into high value-added biobased products, the challenges for a successful industrial implementation, the associated benefits, and some future directions.

Potential benefits of UCOs as oleochemical feedstock

The main raw materials of the oleochemical industry are vegetable oils and animal fats, with the former having a 99.9% share in volume and the remaining small fraction mainly corresponding to butter, fish oils, and fats from animal rendering [16]. Figure 1 presents the historical production of vegetable oils and the corresponding distribution regarding final use. As observed, current production borders 210 Mt/yr., from which 140.8 Mt (i.e. ~67%) are used for food applications (i.e. cooking oils, food ingredients) and 29.8 Mt (~ 14%) used in biofuels, mainly biodiesel production (~1.1 kg_{Oil}/1 $kg_{Biodiesel}$). The remaining 40 Mt (~ 19%) are destined for feed and other oleochemical uses including drop-in applications (e.g. additive for polymers, resins, asphalt, lubricants, greases, drying oils, rubber products, and so on) and as feedstock for different chemical derivatives. Taking into account the estimated global UCO generation (41 Mt/yr., [9]), this amount can replace a large fraction of the virgin vegetable oil currently used as feedstock for the oleochemical industry, a part of which is used as a biodiesel feedstock. Hence, the exploitation of UCOs as chemical feedstock within a circular economy model would help to reduce the environmental and social impacts associated to both, the edible oil and the oleochemical industries.

Figure 1



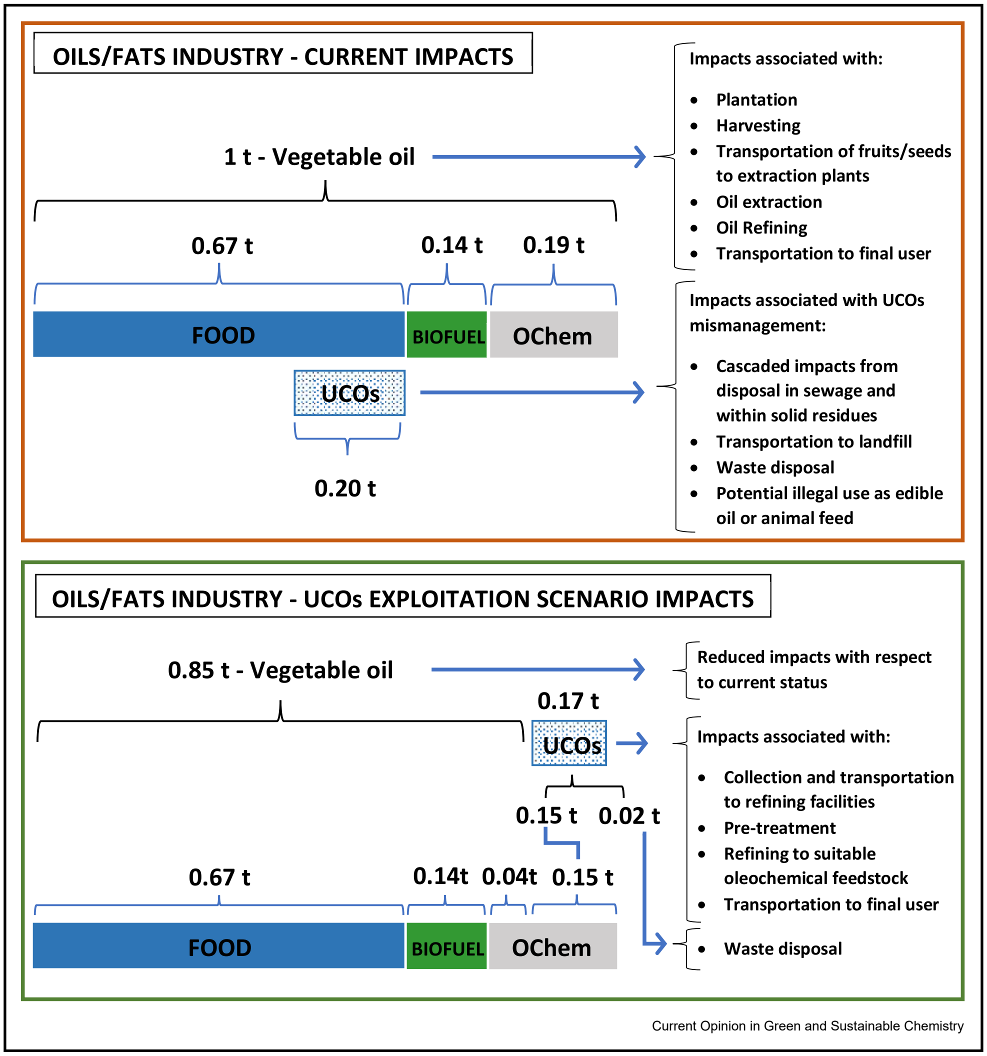
Historical world production of vegetable oils and share among final uses [16].

Figure 2 schematically presents the different stages in which negative social and environmental lifecycle impacts could be mitigated by using UCOs as oleochemical feedstock [17–22] This plot is constructed with reference to each ton of vegetable oil used in the lipids and oleochemical market. It was also developed on the assumption of a 100% efficiency in UCOs collection and reuse, and considering the more conservative estimations of UCOs generation with respect to vegetable oil consumption (20%, [9]). Nonetheless, it is important to point out that for instance in the European community, only 45-50% of collectable UCO is recovered, with a best-case scenario of 70% [11,23]. As mentioned, according to rough estimates, around 0.20 t of UCOs are produced per 1 t of vegetable oil consumed in the market. Therefore, the life cycle impacts of each ton of produced vegetable oil in the oleochemical chain (i.e. agricultural stage, extraction, oil refining, transportation, final use, etc.) adds to the impacts of the produced UCOs (e.g. waste mismanagement impacts, transportation, emissions from landfill and wastewater plants, etc.). Alternatively, using the same 1-ton reference for the total oleochemical market, and maintaining the fractions of vegetable oil used for food (i.e. 0.67 t), biofuels (i.e. 0.14 t) and oleochemicals (i.e. 0.19 t), it would be possible to reduce the required production of virgin oil to 0.87 t by using a circular economy approach. In this case, the production of UCOs would correspond to around 0.17 t, that after pretreatment and refining (i.e. with conservative yield of 90% [19]), it would generate around 0.15 t of UCOs suitable as oleochemical feedstock. Then, the refined UCOs could be used in the manufacture of biofuels, but preferably in value-added oleochemical applications. Whit this approach, the life cycle impacts associated to the global production of vegetable oils would be reduced around 17%, the impacts of UCOs generation mismanagement would shrink, and additional benefits would be obtained if the renewable oleochemicals are used to replace fossil-based products.

High value-added application for UCOs

The valorization of UCOs via transformation into suitable oleochemical products has captured the attention from academic and industrial researchers during the last two decades. Figure 3 presents the evolution of the scientific production (i.e. articles and patents) dealing with the exploitation of UCOs. While the studies on biodiesel are still predominant, there is an increasing trend to explore novel applications, mainly focused on value-added products. In addition to the availability of financial resources, most research studies have been conducted in countries where UCO mismanagement can be a major problem, either because of their large

Figure 2



Potential impact mitigation of using a circular economy approach in the exploitation of UCOs as oleochemical (OChe) feedstock considering life cycle stages (Cradletogate). Functional unit: 1 t vegetable oil. Assuming a 90% yield of refined UCO from the collected one. UCO, used cooking oil.

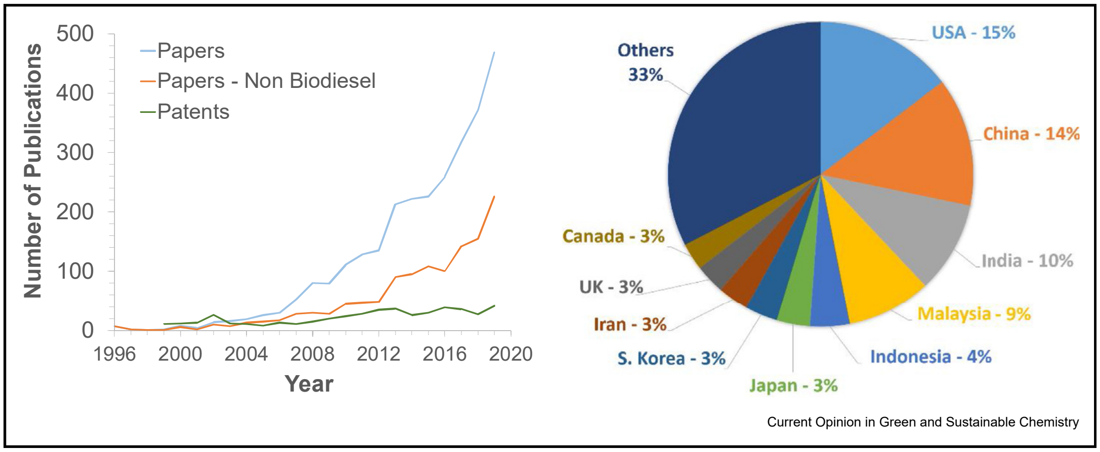
population (e.g. China, India) or for the large per capita generation (e.g. USA, Indonesia, S. Korea). Most EU countries are grouped as ‘others’, and in this case, their large scientific productivity has been promoted by the public policies of the community [20].

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

Challenges and future directions

As observed, UCOs can be used as raw material for a large variety of green chemicals. In addition to the technical limitations observed in some of the processes, there are a number of issues that need to be overcome to enable a succesful industrial implementation. As in any other biorefinery, the supply chain plays a major role in the sustainability of the proposed production schemes. In this case, a major fraction of the generated UCOs

Figure 3



number of publications and patents on the valorization of used cooking oil in the last decades and share by major contributing countries (March, 2020). Searching terms: TITLE-ABS-KEY ("Used cooking oil" OR "Waste cooking oil" OR "Yellow grease" OR "Brown grease" OR "Trap grease") (Source: [24–26]).

Table 1

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

Application	Process	Product	Chemistry behind product use	Highlights	Ref.
UCO valorization					
Plasticizer	Transesterification of UCO biodiesel with 2-ethylhexanol and further epoxidation	Epoxidized 2-ethylhexyl fatty ester (Ep-WCOEtHEs)	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 thermooxidative 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_2SO_4 as catalyst	[37]
	Esterification and transesterification with methanol and aminomethylation (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	[39]

Table 1 (*continued*)

Application	Process	Product	Chemistry behind product use	Highlights	Ref.
				of the asphalt binder are improved	
	Drop in. 0.4–0.8% wt. addition	Macadam pavement	UCO physically cover aggregates	Improves cracking and fatigue resistance	[40]
	Copyrolysis of UCO with rubber	Rubber/UCO binder	UCO reacts with rubber polymers during desulfurization and pyrolysis	Improved low temperature properties of binder Improved rheological properties of asphalt	[41,42]
	Drop in 5% wt. addition	Binder replacement	Unsaturation 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 improved the durability performance of asphalt mixture.	[45]
Masonry binder	Drop in 10% wt. addition	Construction block	Oxypolymerization and cross-linking	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 biofoams can be produced by replacing 40–60% of polyol with biobased alternatives	[48–50]
Lubricant	Epoxidation and hydroxylation with methanol, ethanol, and 2-ethyl hexanol, esterification with hexanoic anhydride	UCO and UCO FAME poliol hexanoic ester	Enhanced thermooxidative 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 thermooxidative 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]

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Table 1 (*continued*)

Application	Process	Product	Chemistry behind product use	Highlights	Ref.
	Drop in 15% wt. addition	UCO dispersible Cu nanoparticles	Unsaturations complexed on the nanoparticles surface.	Particles are synthesized in UCOs 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 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 comprises 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 *Cupriavidus necator* H16 compared with 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 18,342 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	[63]
	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 a superior substrate than glucose but low titers obtained 2.5−2.7 mg/L	[65]

Table 1 (*continued*)

Application	Process	Product	Chemistry behind product use	Highlights	Ref.
Structured materials	Double thermal chemical vapor deposition	Graphene	Pyrolyzed 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 cross-linking 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 a 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 micromesoporous carbon nanocasted on HZSM-5/SBA-15	Carbonaceous material contains oxygen-rich groups suitable for adsorption of cationic dyes. Ordered micromesoporous structure combines size selectivity and high diffusion rates	Material exhibits high adsorption capacity comparable with activated carbons	[70]
UCO-based glycerol valorization					
Carbon source	Fermentation	Hydrogen	Biobased building blocks	Bioconversion of crude glycerol by subtropical mixed and pure cultures. 15.14°moL°H_2/mol glycerol	[71]
		Lactic acid		Bioconversion to lactic acid by *Rhizopus microsporus*. 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 *Klebsiella pneumoniae 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	[75]

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Table 1 (*continued*)

Application	Process	Product	Chemistry behind product use	Highlights	Ref.
				low ethanol addition (15% of the glycerol-COD)	
		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 *Trichosporon oleaginosus*. 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 *Candida zeylanoides* 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]

comes from the household segment, for which very low recovery efficiencies are typical (<6% [11]). Hence, it is necessary to deploy effective policies and regulated practices to enhance UCO recycling and collection rates, under multistakeholder considerations (i.e. authorities, generators, collecting companies, biorefineries). This also indicates that there is need to optimize the collection schemes to ensure that the consumed resources (e.g. energy, financial) do not surpass those from the obtained UCOs, mainly when the biorefineries operate as centralized facilities far from the source. For instance, one study has shown that from a life cycle perspective, biodiesel from European rapeseed UCOs is less sustainable than petroleum diesel and even less than biodiesel from Indonesia's palm oil UCOs [18].

Another major challenge is the highly heterogeneous nature of UCOs. They exhibit a large variability in physicochemical and sensory properties and a substantial amount of impurities [8,79,80], resulting from different diets, culinary practices, and management procedures. In most of the studied processes of Table 1, there were reports of impurities in the raw material affecting the catalytic and biologically conversion steps, the drop-in uses, and even the thermochemical transformations. In addition, unpleasant sensory properties (e.g. color, appearance, odor) are of major concern. Therefore, suitable upgrading processes must be implemented to enable the efficient transformation of UCOs to the desired biobased chemicals without compromising the economic feasibility. Currently, treatment via adsorption with active materials (e.g. silica, clays), liquid-liquid extraction with ethanol, water or phosphoric acid solutions, esterification or neutralization of free fatty acids, vacuum distillation for volatiles and moisture removal, and chemical bleaching using oxidizing agents have been used [8], [80,81,86]. Via these physical and chemical pretreatments it is possible to reduce problematic impurities and to obtain refined UCOs suitable as oleochemical feedstock. . In any case, even after pretreatment and upgrading, the potential presence of trace impurities also might prevent that some of the derivatives could be used in sensitive applications (e.g. personal care products, cosmetics, food or pharmaceuticals) where the market is more attractive. Alternatively, they could be directed to other high value-added markets such as construction materials, asphalt, rubbers, lubricants, surfactants, fuels, and so on. In the same direction, resilient and intensified pretreatment and valorization technologies must be also developed to enable harnessing of other types of waste lipids (e.g. trap grease, rancid oils, food/solid waste lipids, etc.).

A current threat to the industrial implementation of UCO-based chemicals is that they are strongly linked to the biodiesel market, and there are some concerns about the sustainability of this fuel especially given the rapid move from liquid fuel to electric-powered vehicles. Nowadays, UCO biodiesel is promoted via public policies such as the Renewable Energy Directive from EU. According to this directive, UCO biodiesel can be double counted, so its price can be higher than first-generation biodiesel, encouraging supply and demand. Nevertheless, recent claims indicate that at least one-third of UCO-based biodiesel in the European market is fraudulent because apparently it corresponds to the recently banned palm oil biodiesel [82]. Besides, some of the unsustainable palm oil that was prohibited in the EU has been diverted to China for animal feed to replace the UCOs that are currently exported to Europe [83]. These types of problems might push for revisions of Renewable Energy Directive, which will directly affect UCO supply for the oleochemical industry [84]. Finally, the current coronavirus disease (COVID-19) pandemic is putting pressure on UCOs' global trading, affecting supply, dropping prices, and reducing the potential profitability of the biorefineries [85].

Concluding remarks

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

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.

Acknowledgments

This work has been partially funded by the Royal Academy of Engineering under the grant IAPP18-19\65 and the project title: *Valorization of Urban Used Cooking Oils by transformation into value added oleochemicals. Study Case for Bogota, Colombia*.

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* of special interest
** of outstanding interest

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