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Farid Chemat, Maryline Abert Vian, Harish Karthikeyan Ravi

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# 1 **Towards petroleum-free with plant-based chemistry.**

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3 Farid Chemat \*, Maryline Abert Vian, Harish Karthikeyan Ravi

4 Avignon University, INRAE, UMR 408, GREEN Extraction Team, F-84000 Avignon, France.

5 Farid.Chemat@univ-avignon.fr

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## 8 **Abstract**

9 Depletion of fossil resources, global warming, and increasing world's population represents major  
10 Damocles' sword for humanity to ensure its future against famine, climate change and the end of  
11 the petroleum era. Solutions will come from production of plant-based chemicals which is not  
12 new, nor a historical artefact. On earth, 99% of the biomass alive is composed of plants and  
13 microorganisms. Due to this biodiversity, it could be sufficient as a comprehensive sustainable  
14 resource of reagents for industry. Against this background, what will be needed for the industry  
15 worldwide to solve the long-standing unresolved problem of how to convert plant-biomass into  
16 reagents, ingredients and products with acceptable societal, environmental, and cost levels? There  
17 is an urgent need for conceptual leap in fundamental and applied research by original “multi-  
18 scales understanding”. Mitigating climate change fueled by anthropogenic activities with plant-  
19 based green chemistry to establish a circular bio-based economy while adhering to the sustainable  
20 development goals is the scope of this perspective review.

21

22 **Keywords:** Plant-biomass, Reagents, Plant-based green chemistry, Sustainable Development  
23 Goals

24

25

## 26 **Introduction**

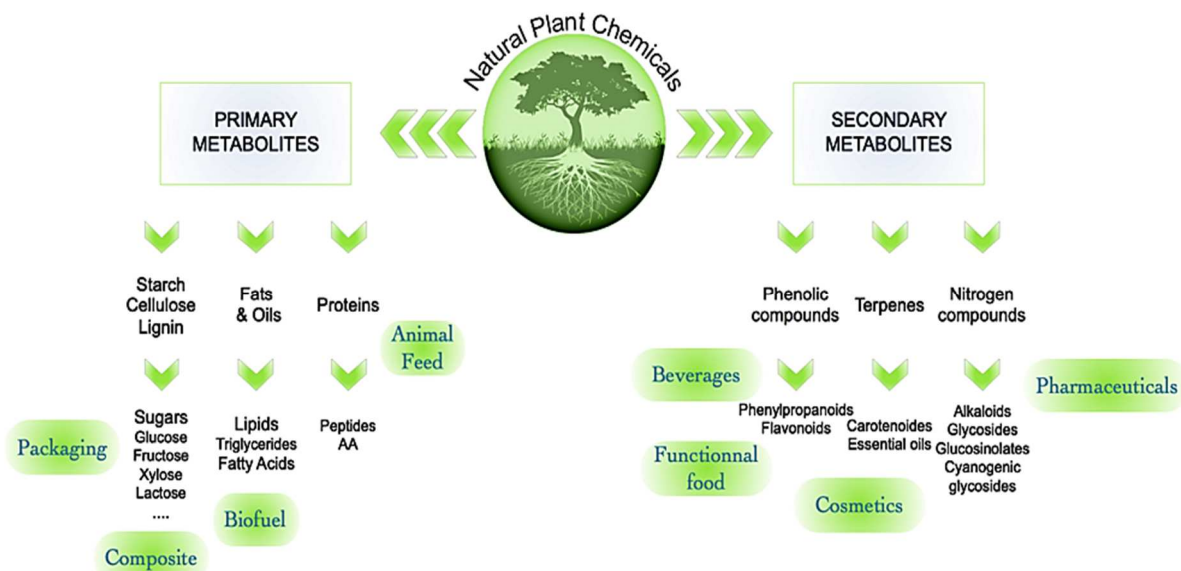
27 Plant-based chemistry has been used probably since the discovery of fire. Egyptians and  
28 Phoenicians, Jews and Arabs, Indians and Chinese, Greeks and Romans, and even Mayas and  
29 Aztecs all possessed a culture of using plants as source of reagents for cosmetic, perfumery,  
30 medicine, food ingredients and products, colors and dyes, and building materials. Until the start  
31 of the petroleum era, plant-derived biomass, due to plant biodiversity, was the primary source of  
32 reagents, ingredients and products for food and non-food applications. The spectacular growth of  
33 petroleum-based processes led to a withdrawal from those based on plant biomass. Almost all  
34 major economies in the developed and developing world have mature refineries which could  
35 transform petrol, a complex liquid mixture to variety of reagents and products essential and vital  
36 for our life and modernity.

37 Depletion of fossil resources, global warming, and increasing world's population represents major  
38 Damocles' sword(s) for humanity to ensure its future against famine, climate change and the end  
39 of petroleum era, which are interconnected. If we consider that these fossil resources have been  
40 formed from a large number of plants, algae, and zooplankton, and also the point that on earth,  
41 99% of the biomass alive is composed of plants and microorganisms, and it is evident that they  
42 could be sufficient as comprehensive sustainable resources for reagents in chemical and food  
43 industries for millions of years. The future of humanity could be secured by establishing a  
44 sustainable and circular economy that relies on the biodiversity with not only plants as green  
45 solution, but also macro and microalgae as blue solution, and microorganisms as white solution,  
46 and insects as brown solution.

47 Nowadays, based on research and innovations in the 20<sup>th</sup> century, we know that in a technical  
48 point of view, almost all petroleum based-chemicals and materials could be substituted by their  
49 plant-based counterparts. However, the cost of bio-based production in many cases exceeds the  
50 cost of petrochemical production. With a petroleum refinery, we can separate and transform petrol  
51 to huge number of alkanes and aromatic compounds as building blocks; in contrast; agri-food  
52 industries largely adhere to the "one cultivation - one product paradigm". The problem is "*the*  
53 *missing link*": how to convert plant-biomass into reagents, ingredients and products for industries  
54 worldwide with acceptable societal, environmental, and cost levels. Solutions require innovations  
55 that break away from the past rather than simple continuity with "*Plant-Based*" Green Chemistry.

56 Green Chemistry is based on principles [1] that could reduce the environmental impact of  
57 nonrenewable resources (petrol, gas and coal) on chemical and food industries: perfumery,

58 pharma, food, fine chemicals, pesticides... Many industries and academia adhere to these  
 59 principles and change the face of chemistry. The next challenge will be the use of starting  
 60 chemical materials from renewable resources such as plants and microorganisms, but also to  
 61 obtain these synthons and ingredients by sustainable, green extraction and separation technologies  
 62 (Figure 1).



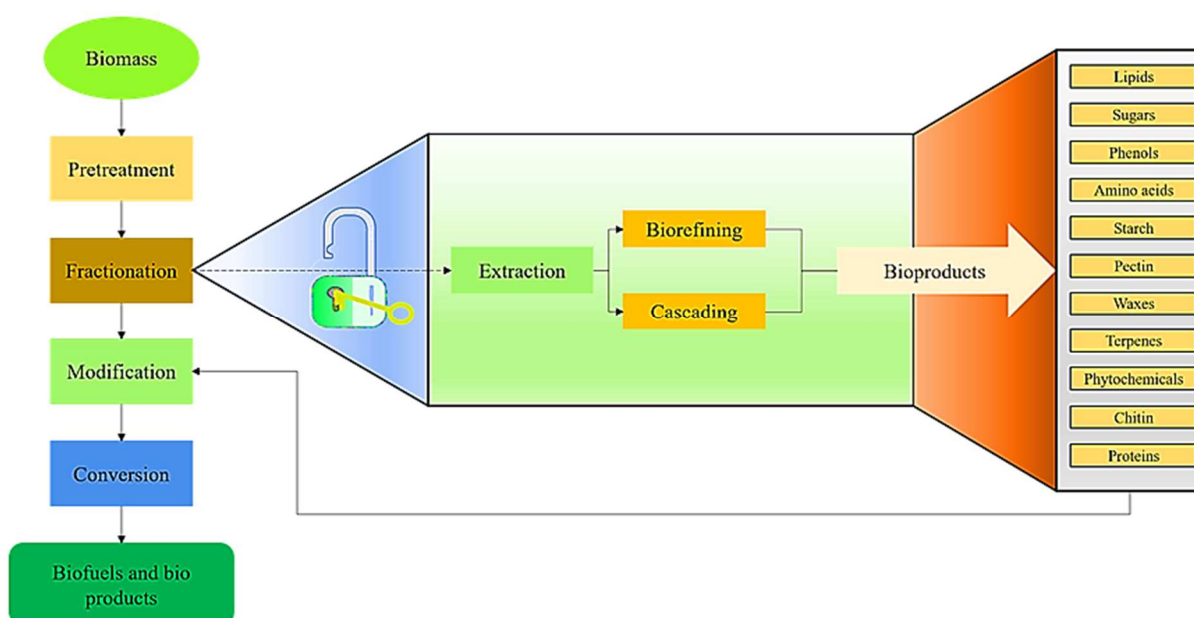
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**Figure 1:** Classes of natural plant chemicals.

### 65 **Extraction: Unlocking the gateway to plant-based green chemistry**

66 The primary raw material for the obtention of petrochemical feedstocks is crude petroleum oil  
 67 which is in turn refined and fractionated as per requirements. The key distinction between the  
 68 refining of petroleum crude oil and biorefining of plant biomasses is in the apparent state of the  
 69 initial material. In the case of crude oil refining, the starting material (i.e. crude oil) is in a liquid  
 70 or slurry state with impurities. Whereas, in the case of plant matrices, the initial biomass  
 71 predominantly exists in solid-state. Benign extraction techniques are the first and foremost step  
 72 employed in order to retrieve the natural plant chemicals such as the primary and secondary  
 73 metabolites. Implementation of a non-destructive extraction technique as the primary step in a  
 74 biorefinery or cascading use scheme would enable further improvement in biomass valorization  
 75 resulting in the obtention of several chemical constituents (Figure 2) including essential oils,  
 76 waxes, sterols, triglycerides, phospholipids, fatty acids, polyphenols, pigments, proteins and  
 77 carbohydrates [2,3]. Extraction process comprises several unit operations namely pretreatment of  
 78 plant material (thermal or non-thermal drying, size reduction...) and post-treatment of liquid  
 79 extract which includes separation, concentration, purification, etc. The principal unit operation in

80 this dynamic is the solid-liquid extraction which is the most common in plant material based  
 81 extraction systems. Optimization of the extraction parameters is crucial and failure to do so often  
 82 might result in time-consuming and energy-intensive processes [4]. The growing importance of  
 83 green extraction and its application for plant-based biomass refinery has been the subject of  
 84 numerous research and review articles [5,6,7,8]. The utilization of bio-based synthons and  
 85 ingredients undeniably offers various advantages but the transformation has to be feasible and can  
 86 not come at any cost [9]. The viability of embracing bio-based products has to be reasonable in  
 87 terms of economy and ecology; that way the sustainability aspect of the transformation process is  
 88 preserved. The ideal scenario is not only to have bio-based ingredients but also to have bio-based  
 89 ingredients extracted with green technologies.



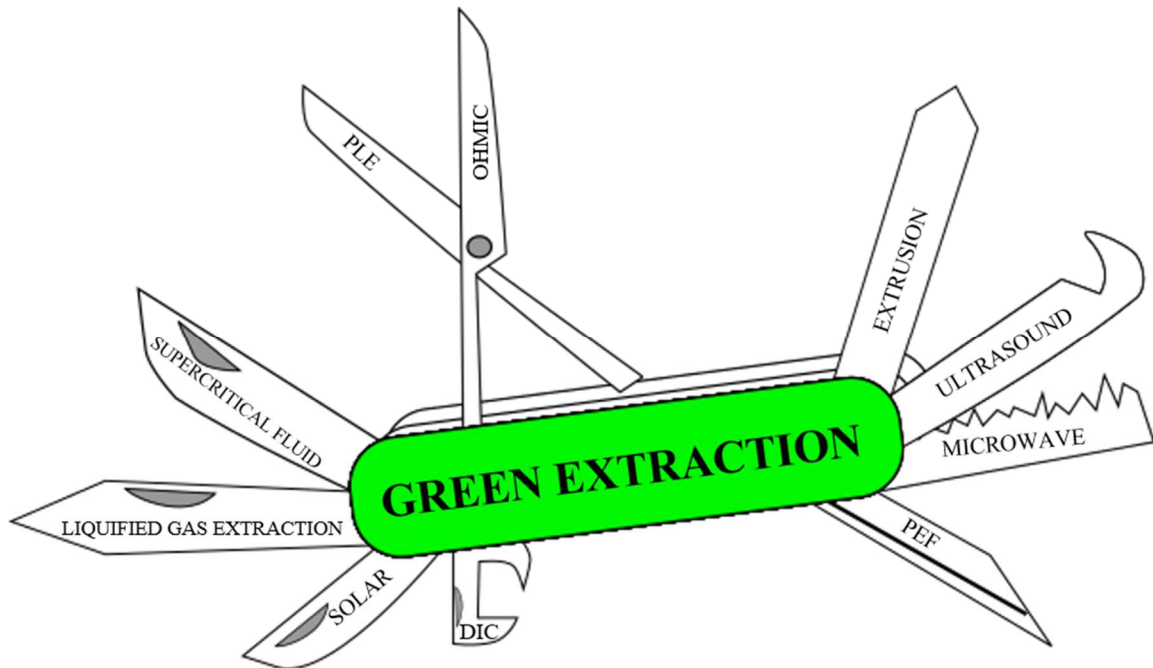
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92 **Figure 2.** Extraction: a key component in plant-based green chemistry.

### 93 **Future challenges: Green intensification techniques and alternative solvents**

94 The major impediments in the transformation of biomass for the provision of chemical  
 95 compounds and bioproducts are the inherent complexity and variation in their physical and  
 96 chemical compositions. This necessitates the adoption of highly complex and tedious processing  
 97 conditions which drives up the operational costs and the low conversion efficiency of biomass to  
 98 products could jeopardize the economic viability of the operation [2,10]. The application of  
 99 innovative extraction technologies and intensification techniques such as ultrasound, microwave,  
 100 instant controlled pressure drop, sub or supercritical fluid, pulsed electric fields, extrusion, ohmic,  
 101 ultraviolet (UV), infrared (IR), and solar-assisted as a standalone process or in synergy can be

102 used for exhaustive recovery of bioactive compounds from plant matrices (Figure 3).  
103 Incorporation of these green extraction and intensification tools considerably enhances the  
104 efficiency, drastically reduces the time, energy and volume of the solvent when compared to the  
105 conventional setup [11].

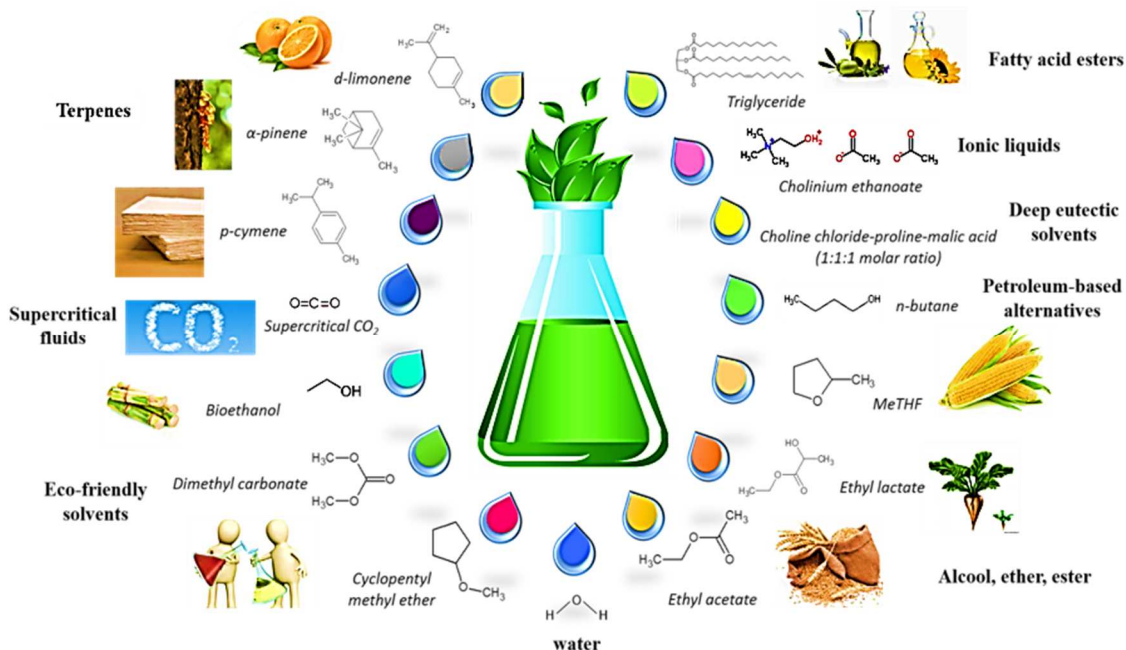


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**Figure 3.** Overview of green extraction and intensification techniques.

108 Finding a suitable alternative to replace petroleum-based solvent for the green extraction of  
109 natural products is an intricate task. The ideal alternative solvent should possess the following  
110 traits: high solvency; high flash points; low toxicity; lower environmental impact; easily  
111 biodegradable; origin from renewable resources, reasonable priced, and easily recyclable without  
112 any deleterious effect to the environment. Several studies focusing on alternative solvents (Figure  
113 4) for the green extraction of natural compounds have been communicated and are summarized in  
114 [12].



115

116

**Figure 4.** Alternative solvents for green extraction

### 117 “One cultivation – multi-product”: A biorefinery paradigm

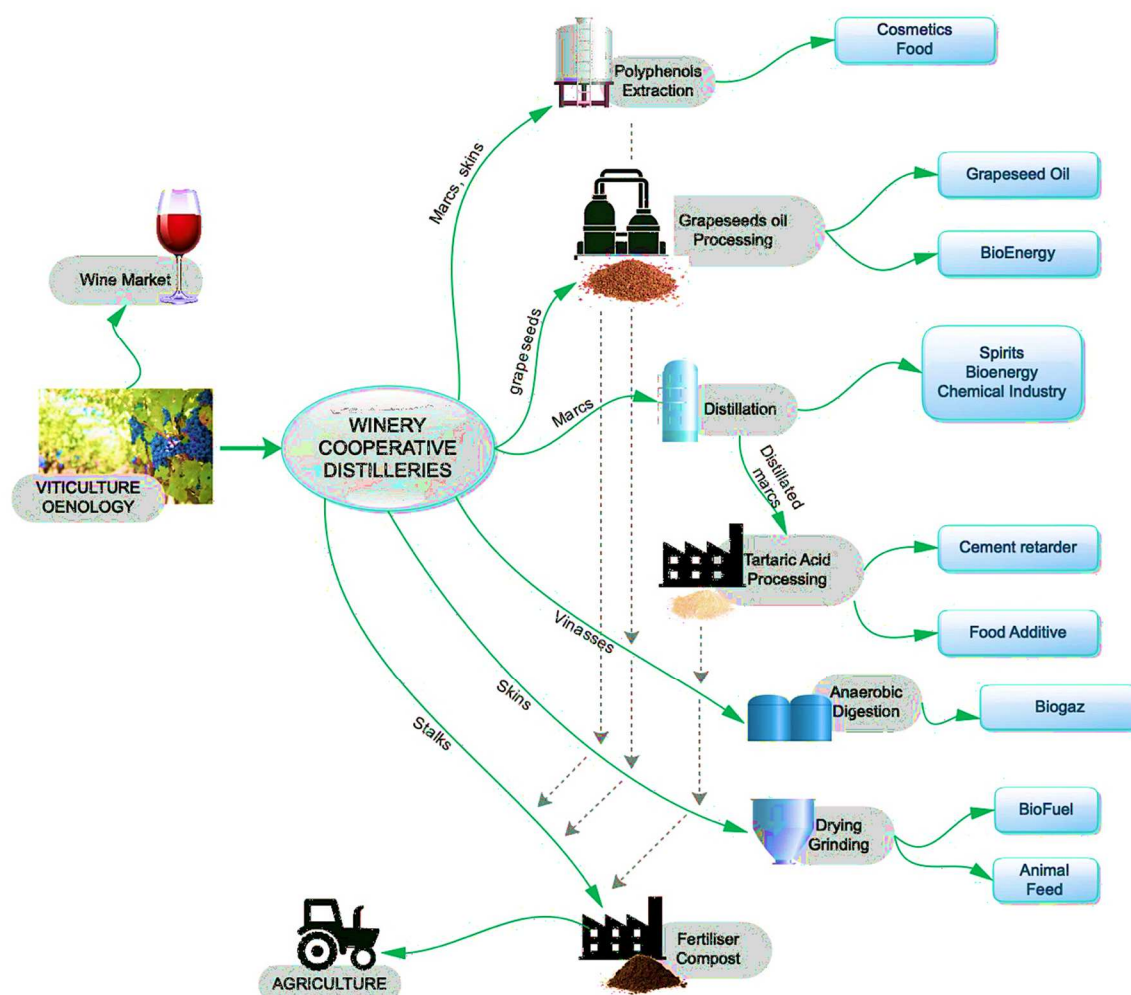
118 Alerts over the last few decades firmly pushed two critical aspects of plant biomass from  
 119 agriculture and food industries largely adhering to the one cultivation-one product paradigm. The  
 120 diminished use of the byproducts because they are dilute resources, and the high cost of recovery  
 121 associated with it are the current obstacles in biomass valorization. The final objective is not to  
 122 provide solutions for a type of biomass, but rather general models applicable to every kind of  
 123 potential biomass substrates. To facilitate the transition of fossil centric global economy to a  
 124 sustainability-based bioeconomy, a forward-looking approach with a stepwise adaptation of global  
 125 refining schemes for the production of bioenergy, biofuels and biobased products should be  
 126 considered. Authors Champagne and Matharu consolidated the processes involved in the  
 127 refinement of biomass to biofuels and bioproducts along the entire supply chain into pretreatment,  
 128 fractionation, modification and conversion routes [13]. Pretreatment processes have ubiquitous  
 129 applications in the conversion of lignocellulosic biomass [14] and can be employed optionally for  
 130 other plant-based biomasses as well. Within this framework, the fractionation process for biomass  
 131 valorization can be categorized into dry and wet processing modes. Fractionation of biomass can  
 132 be accomplished with different strategies: a) Biorefining: The ideal goal of any biorefining  
 133 scheme is to completely valorize the biomass into a spectrum of bio-based products (food, feed  
 134 and platform chemicals) of economic value and exhaust the potential functionalities offered by the  
 135 matrix. This is usually achieved in a single step or multiple steps by employing various unit



136 operations sequentially. b) Cascading use: systematic effort to exploit the biomass for higher-  
 137 added-value products before utilizing it as a source for energy [15]. The deconstruction and  
 138 cracking approach are other plausible alternative strategies for biomass valorization [9].

### 139 **Biorefinerie(s) as success stories for Plant-Based Chemistry**

140 The objective of biorefining is to valorize all the plant's components into products of economic  
 141 significance [16]. At present, there are several types of biorefinery schemes that can be employed  
 142 depending on the different inputs and outputs emerging from the transformation process of plant  
 143 biomass. Almost 80% of the grapes harvested (77.8 million tons in 2018) is used for viticulture.  
 144 The complete valorization of the sheer amount of by-products emerging from winemaking by  
 145 means of an integrated biorefinery approach is an excellent example (Figure 5).



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147

**Figure 5.** Scheme of wine biorefinery.

148 An innovative approach, which seems surprising, is the utilization of insect larvae for the  
 149 bioconversion of different biomasses. Insect-based products for food and feed applications has

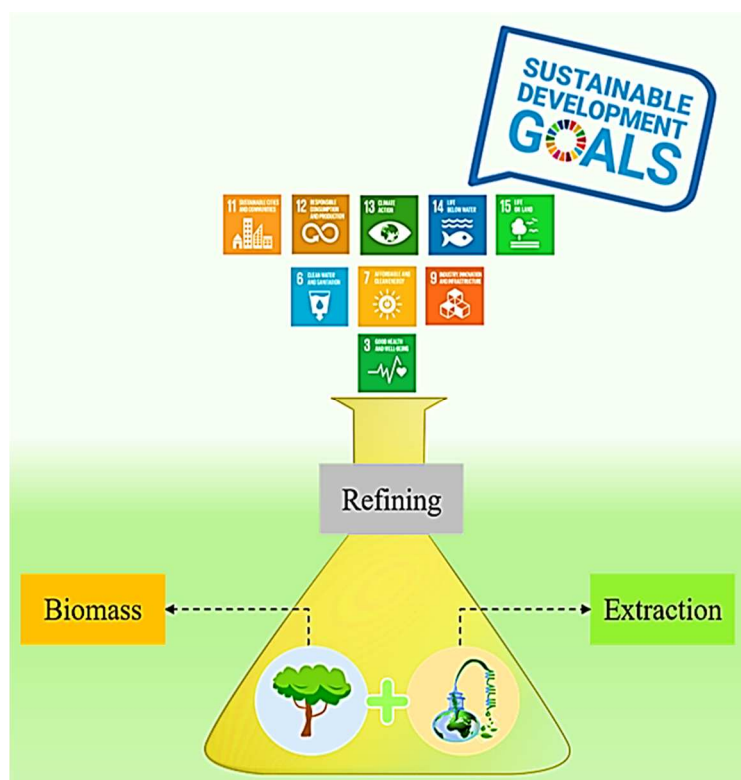


150 already garnered significant attention [17]. They are considered as an alternative to conventional  
151 protein sources like fishmeal and soybean meal. The black soldier fly (BSF) in particular can be  
152 reared on a diverse range of substrates including agricultural wastes and food industry by-  
153 products. Insect bioconversion is exceptionally advantageous in terms of overcoming the physical  
154 and chemical variation challenges in the biomass. Their ability to thrive in a wide array of  
155 substrate with varying nutritional composition can be exploited and used to fabricate a structured  
156 bioconversion system which ensures thorough utilization of the biomass and obtain a standardized  
157 nutritional composition in the larvae. For instance, 1000 kg of fruit and vegetable wastes through  
158 larvae mediated bioconversion yields 125 kg of fresh larvae and almost 250 kg of frass [18].  
159 Though the proximate composition of larvae may vary, the bioconversion ensures assimilation of  
160 high-value components (protein, lipids, minerals and other nutrients) in the larval biomass which  
161 can be retrieved by traditional biorefining or fractionation techniques. The lipid fraction of the  
162 insect finds applications in feed, food, biodiesel, and cosmetic formulations. The protein meal is  
163 widely considered as replacements for conventional protein sources. Chitin has multifarious  
164 applications and frass from insects can be used as fertilizers and soil amendments. Even the  
165 conversion of complex lignocellulosic biomass (corn stover, rice straw) can be effectuated with  
166 insects along with co-conversion agents (microbes, enzymes) [19,20,21]. The conversion  
167 efficiency of lignocellulosic biomass to platform chemical or intermediary products along with  
168 techno-economic analysis and life cycle assessment of the process could be compared with insect  
169 bioconversion for similar biomasses. Such comparisons could shed light on the economic aspects,  
170 environmental impact, technical feasibility, and overall implications of the individual systems and  
171 in turn aid in the industrial-scale adaptation of the process for guaranteed profitability and  
172 sustainability.

### 173 **Plant-based green chemistry: Sustainable Development Goals perspective**

174 The Sustainable Development Goals (SDGs) put forth by the United Nations (UN) is a  
175 culmination of criteria and strategies that serve as the blueprint to achieve a better and more  
176 sustainable future for all. The 17 goals are all interconnected and achieving even one of this  
177 ambitious goal by the year 2030 will have a positive ripple effect on the rest. Adapting green  
178 extraction and refining processes for the production of platform chemicals, energy, biofuels,  
179 active pharmaceutical ingredients and other chemical intermediates will significantly accelerate  
180 the progress towards a sustainable bio-economy. Authors Anastas and Zimmerman addressed the  
181 challenges of sustainable chemistry relevant to SDGs [22]. We postulate that the plant-based  
182 green chemistry approach can have an overall impact on the 17 SDGs and it certainly influences 9

183 of them directly namely (Figure 6), (i) Good health and well-being: replacing petroleum solvents  
 184 with alternative bio-based solvents for example, n-hexane with 2-methyloxolane can change the  
 185 way edible oil refining functions; (ii) Clean water and sanitation: sourcing of biomolecules like  
 186 chitin and chitosan which acts as a flocculant in water treatments, larvae-mediated waste  
 187 management in animal agriculture could drastically reduce the industrial run-offs in animal  
 188 agriculture; (iii) Affordable and clean energy: plant lipids for biodiesel production, biogas  
 189 generation from plant biomass as a last resort in the biorefining scheme or cascading use; (iv)  
 190 Industry, innovation and infrastructure: plant-based green chemistry has already paved the way for  
 191 the implementation of several innovative extraction and processing techniques in industrial-scale.  
 192 Further push by incentivizing the companies that gravitate towards green processes will spur more  
 193 innovation for clean label products; (v) Sustainable cities and communities: self-sufficient  
 194 communities and smart cities with urban vertical farms is a possibility in the near future; (vi)  
 195 Responsible consumption and production: biodegradable plastics from plant-based constituents  
 196 like starch and biopolymers along with regulatory push can boost responsible consumption (vii)  
 197 climate action; (viii) Life below water; and (ix) Life on land: lower greenhouse emissions as a  
 198 result of transformation to plant-based biomass, reduced carbon footprint and sustainable plant  
 199 and animal agriculture practices are few of the aspects that can contribute to the achievement of  
 200 the goals outlaid.



201

202

**Figure 6.** Plant-based green chemistry and SDGs

**203 Conclusions and future prospects**

204 Plant-based green chemistry could be one of the solutions from the past to the future of humanity  
205 focusing on ecologic and economic chemistry, and as a success story of the evolution of green  
206 chemistry in the 21<sup>st</sup> century towards a petroleum-free world. The most critical aspect is not only  
207 to have 100% bio-based product but to obtain 100% “sustainable” bio-based products with net  
208 positive carbon impacts and limiting the use of petroleum solvents and nonrenewable energy.  
209 There are many key challenges and barriers at different levels from microscale (petroleum-free),  
210 mesoscale (detexturation), to macroscale (biorefinery) to achieve a conceptual leap in  
211 fundamental and even in applied research for conversion of plant-biomass to reagents and  
212 ingredients with societal, environmental, and cost impacts to levels acceptable to secure humanity  
213 against the inevitable end of the petroleum era.

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302