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Review

A Review on the Biorefinery Approach and Marketing Strategy of Leafy Biomass

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Abstract: In terms of sustainability, there is a pressing need to evaluate agricultural and forestry leafy biomass that has no current economic value or may pose a threat to the environment. In this aspect, leafy biomass, which represents around 5% of the total tree, can be used for purposes that would be more profitable and ecofriendly. To this end, biorefinery of leafy biomass in the way of extracting valuable health compounds such as phenolics would ensure a valuable societal health ingredient besides alleviating waste disposal problem. However, during the biorefinery process, the primary challenges start with ensuring the feedstock to produce phenol rich leaves extract followed by their isolation and commercialization of this leaves extract. This current review aims to detail the types of biomasses, biorefinery approach of leafy biomass towards the phenolic extraction, leaves commercialization followed by the marketing strategy of the application of phenol rich leaves extract. The outcome of this overview will serve researchers and relevant industries in understanding the process for feedstock collection and the suitable sectors to apply leafy biomass derived healthy compounds.

Keywords: Antioxidant; biomass; biorefinery; by-products; forestry biomass; phenolic compounds

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1. Introduction

Energy production, biochemical waste minimization, availability of nutritious food, and climate change are some of the major concerns worldwide. To feed the increasing world population, on the one hand advanced agricultural practices and postharvest techniques are inevitable, while on the other hand, these increased food, feed, and energy crop productions has resulted in a huge amount of agricultural wastes and by-products [1]. In other words, the production stage represents 24–30% of global agricultural by-products, while post-harvest stage accounts for 20% [2,3]. The other sector of interest is forestry which also produces large amounts of biomass derived from harvestable yield. In total, the global annual generation of biomass waste is in the order of 140 Gt, and this presents significant management problems for industry in terms of cost and environmental effects induced by their disposal [4]. However, with the increase in environmental awareness, there is growing interest in circular economy where these underutilized biomasses can be a rich and cheapest source of several bioactive components viz. polyphenols, carotenoids, flavonoids, anthocyanins, and vitamins. Sustainability and circularity of processes are key words for industrial sector encompassing the reduction of postharvest losses, valorization of by-products, and extraction of bioactive compounds [5]. This review aims to highlight the types of biomasses, biorefinery approach of leafy biomass towards the phenolic extraction, leaves commercialization followed by the marketing strategy of leaves extract application.

2. Biomass

Biomass can be derived from two sectors namely agricultural biomass and forestry biomass. Agricultural biomass can be categorized as (1) agricultural harvest generated by-products, (2) postharvest by-products, and (3) food-processing by-products and wastes. Agricultural biomass including crop stalks, leaves, hays, straws, twigs, and roots (agricultural harvest generated by-products); fruit peels, rinds, pulps, bran, husks (postharvest by-products); pomace, seed/nut shells (food-processing by-products and wastes), among others, are normally discarded or burned (Table 1) [1,3]. Burning organic material emits nitrogen oxides, carbon dioxide, carbon monoxide, sulfur dioxide, lead, mercury, and other hazardous air pollutants which contributes to climate change, being responsible for an estimated 8% of global greenhouse gas emissions [2,4]. It is estimated that 140 Billion tons of biomass are generated each year from agricultural by-products in liquid, solid and gaseous form [5]. On the other hand, forestry biomass includes residues such as stumps, branches, barks, stems, and leaves and processing waste such as logs and sawdust which accounts for around 4.6 Billion tons annually of which 60% goes to energy generation, 20% to industrial 'round wood' and the remaining 20% being primary production loss that remains in-field to decay [6]. Forestry biomass used in power plants to produce energy causes huge increase in carbon emissions compared to coal and other fossil fuels. However, biomasses are underutilized source of material as such there has been little activity focusing on a comparatively 'low-carbon' route for their valorization [4].

Table 1. Biomasses generated from fruit and vegetable processing industry.

Fruit/vegetable	Biomass	References
Carrot	30-40% (pomace)	Kaur et al. [7]
Tomato	3-7% (seed and peel)	Schieber et al. [8]
Apple	11% (seed core and pulp)	Ayala-Zavala et al. [9]
Banana	≤ 30% (peel)	Schieber, Stintzing and Carle [8]
Guava	10-15% (seed and peel)	Selvamuthukumaran and Shi [3]
Mango	11% (peel), 13.5% (seed), 17.9% (unusable pulp)	Ayala-Zavala, Rosas-Domínguez, Vega-Vega and González-Aguilar [9]
Orange	66% (peel)	Li et al. [10]
Papaya	8.5% (peels), 6.5% (seeds), 32.1% (unusable pulp)	Ayala-Zavala, Rosas-Domínguez, Vega-Vega and González-Aguilar [9]
Pineapple	13.5% (peels), 14.5% (pulp), 9.1% (core), 14.9% (top)	Selvamuthukumaran and Shi [3]

Regarding biorefinery aspect, several reports have shown that agricultural and forestry biomass can be a potential source of bioactive compounds such as polyphenols which are reported to show several biological activities (anti-allergenic, anti-inflammatory, anti-artherogenic, cardioprotective, anti-microbial, anti-thrombotic, and antioxidant effects). Having said that agricultural and forestry biomass could be a good choice as sustainable and renewable feedstock for the functional, nutraceuticals, and pharmaceutical industries [11–19]. To this end, extraction of bioactive compounds using green extraction methods as a 'low-carbon' route can be very efficient knowing their uprising market demand. For example, based on Grand View Research (2019), the global polyphenols market size was already worth USD 1.68 Billion in 2022 and a Compound annual growth rate (CAGR) of 7.4% is expected to register from 2023 to 2030. According to them, the functional food and beverage industries are the largest sectors of the global market share (> 70%) of polyphenols followed by dietary supplements, cosmetics, animal feed, and others [20]. Hence, biomass processing through green extraction technology would offer double benefit (i) extraction of high value compounds that have market demand, and (ii) reduction of carbon emissions, that is caused by conventional biomass-to-energy conversion plant. In this way, use of biomass as a source of extraction materials will not only benefit the agri-food economy but also would have positive environmental impact.

3. Promising biomass as a part of bio-refinery approach

Leaves could be a promising source to exploit valuable compounds such as phenolics. Different phenolic compounds have been separated from different leaves, for instance, chlorogenic acids and anthocyanin from blueberry leaves [21]; 5-*O*-galloylquinic acid and 5-*O*-galloylquinic acid from mastic tree leaves [22]; phenolic acid derivatives and flavonoids from moringa leaves [23]; oleuropein from olive leaves [24]; isoquercetin, catechin, hydroquinin, gallic acid, tannic acid, and rutin from cashew leaves [25], among others. Other biomass of agricultural origin, in specific, biomass form postharvest by-products, and food-processing by-products and wastes, are not homogenous and may come from different fields with differing cultivation procedures, conditions and processing; consequently, a standardization process is necessary. Moreover, agricultural food by-products are characterized by their biological instability due to their high microbial load that can affect product safety and naturally accelerate microbial degradation [5]. Another point is, most of the agricultural biomass coming from processing industries undergo several processing treatments such as heating, which might have negative impact on the stability of phenolic compounds. On contrary, leaves can be a potential source of phenolics and the justification behind choosing leaves among other biomasses of agricultural and forestry origin can be given as such,

- leaves are a sustainable source where overexploitation is believed not to be an issue as leaves can be easily grown back by the plant [26,27].
- leaves can be used as fresh, therefore, can preserve the quality of phenolic compounds.
- leaves can be a cheapest source for the extraction of phenolic compounds.

Note that, leaves from one source might be rich in some selective phenolic compounds, while other leaves may contain higher amount of other phenolic compounds, as phenolic compounds possess a broad category of chemical compounds [28]. To date, more than 10,000 phenolic compounds have been identified in plants [29]. In this scenario, the combination/blend of several leaves extract would offer higher benefit. However, before commercialization of leaves' phenolic compounds (either single/ blend of leaves), thorough toxicological study would be the mandatory step, as leaves may contain toxic substance or potentially toxic substance (that comes into the extract during extraction) [30–32]. Moreover, toxicological study on blend of extract (which is separately safe) need to be performed as well, as there might be any synergistic event that could develop new toxic substance, even though they were not present before the mixing or blending of the leaves / leaves' extracts.

4. Leaves commercialization

Availability of raw material is the prerequisite of any industrial production process that takes part in a circular economy. Likewise, the regular supply of leaves (as raw/semi-processed) throughout the year should be ensured. However, leaves are identified as perishable commodity (moisture content up to 80% w.b.) and hence proper storage is required until they are used for required purposes, for example, extraction, etc. [33]. High water containing commodities are highly perishable as they are prone to microbial growth and enzymatic activity. Therefore, perishable commodities are preferably stored at lower temperatures to increase shelf life and retain quality. For prolonged storage and maintenance of the quality of biological samples, storage at -20°C or lower is preferred. However, storage of fresh biomaterial consumes space and is more expensive in terms of transportation (due to the greater mass from the high-water content). To this context, drying can reduce the space requirements and make the transportation of the dried commodity easier because it decreases the mass of the commodity to be transported, along with increasing its shelf life [34].

Drying is a process to remove moisture from fresh commodities and reduce its water activity, which inhibits microbial growth and minimizes deteriorative biochemical reactions. It also reduces the weight and volume of the sample thereby reducing storage and transportation costs as already mentioned. In addition, drying can modify the physical microstructure of plant tissues, which leads to increased extraction yields [35]. For example, freeze drying causes significant changes in the microstructure of the final dried product making it more porous so that solvents can easily penetrate the sample and thus extract more phytochemicals [36–39]. Several drying methods have been practiced with leaves aiming for extraction of phenolic compounds such as freeze drying [40–47], microwave drying [34,40,42,44,48–50], oven drying [41,44,49], spray drying [51], sun drying [40,42,44], vacuum drying [40], shade drying [43], among others.

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As seen above, for sample preparation for phenolic content analysis, freeze drying has often been applied, however, it is a relatively expensive drying method. For economical commercial applications, the most practical method is hot air drying, however, application of higher temperatures can lead to degradation of the phenolic components [40]. Considering these aspects, it is necessary to identify the most suitable drying method and conditions for a specific type of plant sample. Next to drying, the grinding of dried leaves followed by sieving and storing in controlled environment (temperature, relative humidity, light, oxygen, etc.) would offer rapid industrial processing of maple leaves as their raw material.

To give an example of how leaves can be commercialized throughout the year, here the species *Acer saccharum* is considered as a representative. In Canada, sugar maple leaves calendar year starts in late spring (May) and ends in late fall (November) where they fall to ground to prepare for the winter. Having said that, supply of maple leaves within these months (May-November) can be easily secured. Collecting of leaves in peak season followed by their preservation might also ensure the availability of maple leaves in rest of the months (December-April). Maple leaves can be obtained from both cities and forests. In this aspect, existing maple farms utilized for maple sap collection for the production of maple syrup, could be a good choice to have homogeneous leaves (i.e., maple leaves only). Currently there are 5340 maple farms exist in Canada, with 4776 maple farms originating from Quebec, 391 from Ontario, 111 from New Brunswick, 48 from Nova Scotia, and 14 maple farms from rest [52]. As these farms are under regular maintenance, collecting leaves from these farms besides forests and cities would be most convenient. Maple leaves could be collected from tree/surrounding ground, depending on the time of the life cycle of the tree in a calendar year. Afterwards, the mass-collected leaves would be sorted out (based on shape/density/color/etc.) to separate the targeted leaves from unwanted substances i.e., from damaged and unhealthy leaves, rocks, debris, etc. The sorted leaves could be blown by cold air to remove dirt/debris followed by washing them in running tap water. Then the selected healthy leaves will undergo drying process and followed by grinding and sieving of dried powder. The dried powder will be packed in a good quality container (that would not allow interference from surrounding environment to the leaves powder), and the packed powder will be stored in controlled environment until its industrial processing.

5. Marketing strategy of phenol rich leaves extract

Phenol rich leaves extract obtained through green extraction can be utilized in different sectors based on their functionalities. Knowing that there is already an increasing market demand for phenolic compounds. For instance, Belwal et al. [53] reported that the nutraceutical market share would increase through phenol derived biological materials. In this aspect, the demand for phenolic compounds in global markets was expected to reach USD 873.3 million in 2018 based on the study conducted by Transparency Market Research [54–56]. Therefore, it can be assumed that leaves extracts have a better future to dominate the phenolic market as well as relevant industrial sectors. However, marketing strategy of leaves phenolics could be linked with their several biotechnological activities [28]. Generally, the most analyzed property of phenolics is their antioxidant activities, which have been often associated with the prevention, modulation, or treatment of significant diseases and health disorders in humans, including neurodegenerative and cardiovascular disorders, atherosclerosis, and diabetes [57]. Phenolic compounds are considered as antioxidants due to the donation of a hydrogen atom and/or an electron to free radicals, causing the break of chain reaction of oxidation. The antioxidant effect depends on the number and position of the hydroxyl groups [27]. Other important bioactivities for maintaining good health have also been associated with these compounds, e.g., anti-inflammatory, antimicrobial, and anti-proliferative activities, and these biological activities have aroused interest in the use of these molecules in the formulation of nutraceutical products [28]. In addition to the pharmacological interest in these compounds, their biological activities (antioxidant, antimicrobial) could also be explored in other industry sectors, such as food industries, cosmetic industries, and packaging industries. To be precise, depending on the biological activities showed by phenolic compounds, they can be marketed to the focused industries. For example, phenolic compounds can be used in nutraceutical industries as therapeutic agents; in food industry as food additives (in the form of preservatives- antioxidant and antimicrobial actions; food colorant) and functional ingredients; in packaging industries as active packaging (antioxidant and antimicrobial action) and smart packaging (color indicator for deterioration); in cosmetic industries as antimicrobial, antioxidant, tyrosinase inhibitor, UV protector [28].

5.1. Nutraceutical industries

"A *nutraceutica*l is a product isolated or purified from foods that is generally sold in medicinal/supplemental forms not usually associated with food. A nutraceutical is demonstrated to have a physiological benefit or provide protection against certain chronic diseases" [58]. However, a major challenge of the current nutraceutical sector is ensuring bioavailability for obtaining the desired effect. To define, bioavailability is the fraction of a nutrient that has been digested and absorbed and is available for the metabolic functions of the organisms [59]. Bioavailability can be increased by increasing the fraction/quantity of these compounds or the quality of the extract. In this aspect phenolic nanoparticles can be more effective. Yehia et al. [60] produced phenolic nanoparticles from eggplant skin through dispersion with the application of ultrasounds and obtained higher antimicrobial activity. An enhanced delivery system using nanocarriers can contribute towards increasing the efficiency of the antioxidant [57]. Selivanova and Terekhov [61] has presented the increased bioavailability of flavonoids achieved through the application of crystal engineering which is "an applied field of supramolecular chemistry including design and synthesis of new crystalline compounds with specified physicochemical properties".

5.2. Cosmetic industries

The defensive properties of the phenolic compounds have been useful in other applications. Phenolic acids are being used as natural antioxidants in the cosmetic industry, as they possess defensive properties against growth and evolution during the pathological conditions associated with oxidative stress [57]. Ma et al. [62] evaluated the use of glucitol-core containing gallotannins (GA) of red maple leaves extract in cosmetic applications and found that GA was able to: (1) reduce the levels of reactive oxygen species, (2) down-regulate the expression of MITF, TYR, TRP-1, and TRP-2 gene levels in a time- dependent manner, and (3) significantly reduce protein expression of the TRP-2 gene. Olive leaves extracts had a strong potential as an antiaging ingredient and cosmetic/food preservative exhibiting antioxidant activity, capacity to inhibit elastase (82.5%), collagenase (98.7%) and tyrosinase (50%) [63].

5.3. Food industries

5.3.1. Functional food

According to Health Canada, "A *functional food* is similar in appearance to, or may be, a conventional food, which is consumed as part of a usual diet, and is demonstrated to have physiological benefits and/or reduce the risk of chronic disease beyond basic nutritional functions" [58]. To date, as a source of phenolic compounds, the application of rosemary extract, anthocyanin, steviol glycosides as food additives has been approved by European Union and therefore they are practiced on a variety of food products [64–67]. Due to their strong antioxidant properties, olive leave extracts were seen in pork patties, sunflower oil, palm oil, and olive oil applications [68], *Gingko biloba* leaves extract in pork meat [69], green tea phenolic extracts have been seen in bread applications [70].

5.3.2. Preservatives

Phenolic compounds with strong antioxidant properties can also be used as preservatives. According to the United States Food and Drug Administration (USFDA) Code of Federal Regulation, "antioxidants are the substances participates in physiological, biochemical, or cellular processes that inactivate free radicals or prevent free radical-initiated chemical reactions". Apart from the consumers increasing demand for "clean label" natural ingredients, the raising safety concerns about synthetic antioxidants (e.g., BHA, BHT, PG, and TBHQ) also necessitate the search for effective antioxidants from natural sources [71]. Phenolic compounds are classified as primary antioxidants that delay or inhibit the initiation step or interrupt the propagation step of oxidation by scavenging the free radicals. The antioxidant potential of phenolic compounds however mainly depends on the number and arrangement of the hydroxyl groups (-OH) in the molecules of interest [72]. The phenolic extracts obtained from fermented rice bran were applied in pizza doughs stored at room temperature, where the shelf life increased by 10 days [73]. In a different study by Christ-Ribeiro et al. [74], the efficiency of antifungal effects of phenolic extracts obtained from fermented rice bran and Spirulina sp. have been found to be 2.5- and 1.5-folds, respectively, compared with calcium propionate. Ab Rashid et al. [75] employed microencapsulated forms of anthocyanins from *Clitoria ternatea* as a biopreservative and coloring agent for baked food products, where encapsulation enhanced the stability of the compounds for pH fluctuations and applicability. Moreover, green tea extracts have been used in pork sausages which in turn ensured shelf life extension [76].

5.3.3. Packaging industries

Phenolic compounds have also been proposed as part of the composition of membranes and films packaging material, contributing towards the enhancement of the shelf life of beef [77,78]. Catechin and quercetin as phenolic compounds were used in ethylene-vinyl alcohol copolymer (EVOH) film for the packaging of oxygen sensitive foods, which resulted in the improvement of food stability [79]. Attributing to the sensitivity of anthocyanins to the change in pH, these compounds were utilized for monitoring pH variations on the development of chitosan and starch-based intelligent films embedded with anthocyanins [80]. Gelatin/ polyvinyl alcohol based colorimetric indicator film with mulberry anthocyanins was applied for monitoring fish freshness, where visible color changes were observed with the release of volatile nitrogenous com- pounds over the storage period [81]. Da Rosa et al. [82] developed of biodegradable films with improved antioxidant properties based on the addition of carrageenan containing olive leaf extract for food packaging applications.

5.3.4. Potential application of leaves extracts in food applications

Facing restrictions to the use of artificial food additives, the scientific community has been working on the development of natural alternatives. Moreover, owing to consumers' preferences and demands for functional foods, bioactive phytochemicals from leaves extracts can be progressively applied as the ingredients to improve quality traits, nutritional and therapeutic properties. A list of potential food applications fortified with different plant extracts as a source of phenolic compounds is given in Table 2. Based on the provided food applications fortified with different plant extracts, it can be hypothesized that application of leaves extracts might also be possible to those food as leaves can also possess similar classes of phenolic compounds. For example, with reference to the approval of rosemary extract by the EU, the main phenolic compound isolated from rosemary leaves extracts include coumaric acid, caffeic acid, gallocatechin, luteolin, and isorhamnetin [83,84]. These phenolic compounds have also been isolated in the extract of leaves from stevia, moringa, sea buckthorn, and pistacia [56].

Table 2. Application of phenolic compounds (PCs) in different foods.

PCs	Source	Product	Effects	References
Phenolic extracts	*Matricaria recutita, Foeniculum vulgare.*	Yogurt	Antioxidant activity	Caleja et al. [85]
	Litchi chinensis	Sheep meat nuggets	Antioxidant activity	Das et al. [86]

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	Rosmarinus officinalis	Cottage cheese	Antioxidant activity	Ribeiro et al. [87]
	Pomegranate peel	Ice cream	Antioxidant and α-glucosidase activities	Çam et al. [88]
Hydroxycitric acid	*Garcinia cowa*	Pasta	Antioxidant activity and improvement of sensory quality	Pillai et al. [89]
Anthocyanins	*Euterpe edulis*	Fermented and unfermented beverages	Improvement of the color	Lima et al. [90]
	Phaseolus vulgaris	Sport beverage	Stable color and fortification with bioactive molecules	Aguilera et al. [91]
	Myrciaria cauliflora	Fresh sausage	Improvement of color, antioxidant and antimicrobial activities	Baldin et al. [92]
	Prunus nepalensis	Yogurt, syrup, hard-boiled candy	Stable color and sensory acceptance	Swer et al. [93]
Catechin and epigallocatechin gallate	*Camellia sinensis*	Low-fat hard cheese	Antioxidant activity	Rashidinejad et al. [94]
Carotenoids and phenolics	Mango peel	Macaroni	Antioxidant activity Enhanced sensory properties	Ajila et al. [95]

6. Conclusion and future directions

Leafy biomass coming from either forest or agricultural practices or from food industries, is rich in many beneficial components. Therefore, in stead of leaving them to become the treat to the environment, biorefineries combining the profitable extraction and production of various nutraceuticals, pharmaceuticals, or natural antioxidant ingredients will not only decrease the environmental pollution but also help the economy by creating new sources of income. However, future research on the life cycle assessment of the biorefinery process of leaves toward the extraction of phenolic compounds is recommended to find out the most cost effective and eco-friendly method is recommended. In addition, research is required on the controlled delivery of the leaves' phenolic compounds both in in vitro and in vivo conditions.

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