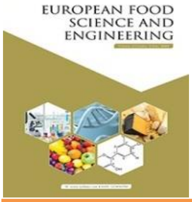

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A waste material rich in bioactive compounds: Hazelnut waste

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ABSTRACT

Nowadays, increasing sensitivity to the environment has led to the development of sustainable agricultural policies. In this respect, it has become important to transform agricultural waste products into value-added products. Hazelnut, which has a significant trade volume worldwide, is processed into products, and some waste materials can be emerge. These waste products could transform into high added-value to food, cosmetics, and pharmaceutical industries due to possessing the bioactive compounds such as phenolics, bioactive peptide and, dietary fibre in them. This review represents the research on the bioactive compounds from the hazelnut waste, especially conducted in recently, and concentrates on its tree leaf, husk, and oil meal.

1. Introduction

Today, the energy crisis in the world and the increasing concerns on the environment have accelerated the research on the search for the alternative energy sources to fossil fuels (Havrysh et al., 2021; Tsekos et al., 2021). One of them is waste. According to Directive 2008/98/CE, '*waste means any substance or object which the holder discards or intends or is required to discard'* (Anonymous, 2008). Based on this definition, waste can be classified as follows; state (solid, liquid, gaseous), source (agricultural, commercial, industrial, municipal), and degradability (biodegradable, non-biodegradable) (Dey et al., 2021; Koul et al., 2022). A considerable amount of waste is generated in the world, for example, only the European Union generated about 2.2 billion tons of waste in 2020 (Anonymous, 2020). On the other hand, in terms of solid waste, 2.02 billion tons of it was produced in the world in 2016, and it is estimated that this number will increase to 2.59 billion tons and, 3.4 billion tons in 2030 and 2050, respectively (Kaza, 2018). If the waste cannot be controlled, that is, if zero waste management is not implemented, the world may inevitably face serious problems. It might affect on both environmental safety by causing air pollution, water pollution, soil contamination and increasing greenhouse gases, and human health (Anonymous, 2016). Due to these adverse effects, the principles of zero waste management have been focused all over the world in recent years, comprising prevention, reduction, recycle, recovery, and disposal (Anonymous, 2008). Therefore, for the implementation of zero waste management, recently, there has been conducted a lot of research on the reusing of the waste from applied as the fertilizer and animal feed (Chew et al., 2018) to manufacturing bio-based films for food packaging (Bastante et al., 2021) and producing biogas (Havrysh et al., 2021), and biodegradable polymers (Maraveas, 2020). Probably, it can seem like that recovering biochemicals, producing energy or value-added products from waste rather than their disposal will be an important philosophy of this century.

Agriculture provides an important source of raw materials for the food industry and human need, and eventually a large amount of agricultural waste is occurred as a result of agricultural and agro-industrial activity (Sharma et al., 2022). Agriculture waste is made up field residues (i.e stems, seeds, husk, shell and so on), industrial processing waste (i.e pomace, sugarcane bagasse, hazelnut cake etc.), livestock waste (bedding/litter, wastewater in the slaughterhouse, animal carcasses etc.), and chemical waste (pesticides, insecticides, and herbicides etc.) (Dey et al., 2021). They can comprise a number of bioactive chemicals such as polyphenols, and dietary fiber, especially in field residues and industrial processing waste (Balasundram et al., 2006; Beutinger et al., 2020; Castrica et al., 2019; Dey et al., 2021). The former has antioxidant, antimicrobial, anticancer, and antiproliferative activity (Beutinger et al., 2020; Castrica et al., 2019; Dey et al., 2021). The latter, which is an essential part of the plant cell wall, may be divided into two subgroups; soluble and insoluble in water (Gill et al., 2021). They have a favorable impact on human health, regulating the bile salts, improving the fecal volume by holding the water, influencing the microbial spectrum to beneficial ones, taking part in the production of short-chain fatty acids (propionate, butyrate, and acetate) in the gastrointestinal tract, which have an important role in both energy metabolism, host immunity and inflammation, and in inhibiting colon cancer cell proliferation (Capuano, 2017;

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Holscher, 2017; Morrison & Preston, 2016; Yao et al., 2022). Because of these positive effects on human health, today, it is recommended to consume 14 grams of dietary fiber per 1000 kcal daily (Anderson et al., 2009). Dietary fiber has been isolated and characterized from various agricultural wastes, for instance, cellulose from waste of wheat straw (Bian et al., 2019), rice husk (Ragab et al., 2018), onion and garlic (Reddy & Rhim, 2018); hemicellulose from rice straw and its husk (Ragab et al., 2018), pineapple peel (Banerjee et al., 2019); lignin from hazelnut and walnut shell (Gordobil et al., 2020); and glucans and pectin from walnut green husk (La Torre et al., 2021).

Hazelnut is one of the nuts, derived from the tree, a genus (*Corylus*) belonging to the Betuliaceae family. It was produced approximately 963 million tons in between 2016-2020 (Anonymous, 2023). Turkey is an important producer, which met about 63% of the world hazelnut production in between 2016-2020, followed by Italy, Azerbaijan, and others (Anonymous, 2023). It is a product with a significant commercial value in the world, mainly for Turkey. The world export value of hazelnut was about 2.2 billion dollars, which of Turkey was 1.3 billion dollars in 2021 (Anonymous, 2023). In addition to its economic value, it can be added to baklava, cakes, ice cream, chocolate, confectionery, cacoa-hazelnut cream (Baycar et al., 2021; Dervisoglu, 2006; Ermis & Ozkan, 2021; Gonzalez-Estanol et al., 2022; Guiné & Correia, 2020), its milk (Gul et al., 2017) and fermented their products (Atalar, 2019; Ermiş et al., 2018) to the alternative commodity to milk, and thus both contributing taste-aroma to the products to which it is added and enriching them in terms of nutrients such as protein (Muller et al., 2020), lipids (Granata et al., 2017), vitamins (Stuetz et al., 2017), minerals (Muller et al., 2020), dietary fibers (Tuncil, 2020) and bioactive compounds (Gültekin-Özgüven et al., 2015; Taş & Gökmen, 2015).

After the hazelnut with green husk is harvested, it is dried in the sun to certain moisture content, and then hazelnut in shell is separated from the green husk by a hazelnut threshing machine. Besides, during this process, rotten and damaged hazelnuts are relatively removed from the healthy hazelnuts with the help of ventilation. Then, the hazelnuts are sent to the factory to be processed into products such as natural hazelnut kernel, roasted hazelnut kernel, chopped hazelnut kernel, hazelnut puree, and hazelnut oil. In the view of the process from harvesting to consumption, there are five hazelnut by-products including hazelnut tree leaf, green leafy husk, shell, skin, and oil meal (Figure 1). This review focused on hazelnut tree leaf, husk and oil meal, but not its shell and skin and it has been recommended for further reading on them as follows; Fuso et al. (2021), Alalwan et al. (2022).

2. Hazelnut Tree Leaf

After harvesting, the hazelnut leaves fall off and may become a natural source of fertilizer for the soil owing to its mineral and organic matter content (Öztürk & Tarakçıoğlu, 2016; Wang et al., 2018). Additionally, it possesses the bioactive compounds such as phenolics (Shahidi et al., 2007), diarylheptanoids (Masullo et al., 2015a; Masullo et al., 2015b), taxanes (Ottaggio et al., 2008), essential oils (Najda & Gantner, 2012) and α-tocopherol (Sivakumar & Bacchetta, 2005).

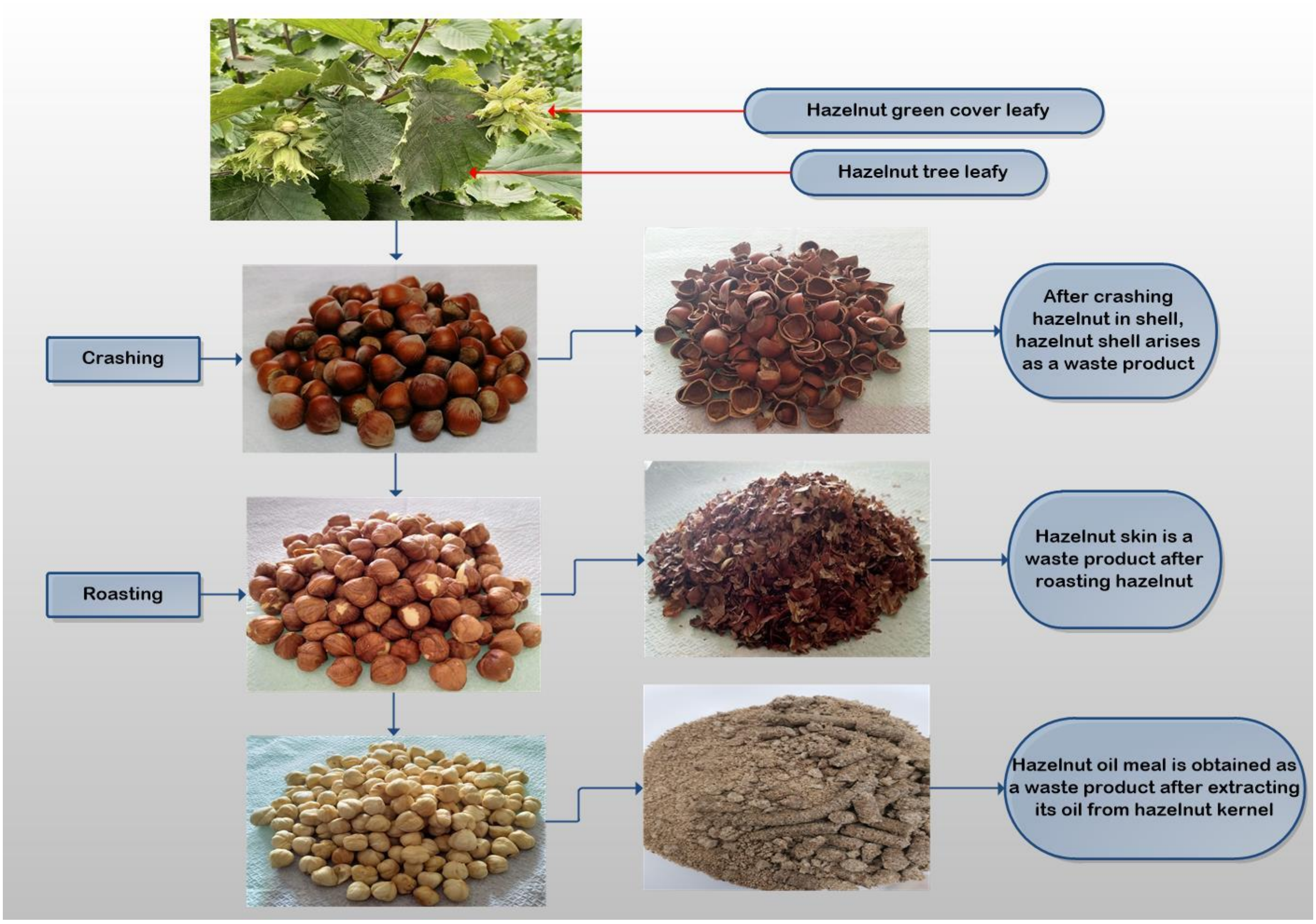

Figure 1. Hazelnut waste

The leaves of the plant may be often used in traditional medicine to treat the swelling, rash, phlebitis, varicose veins, and hemorrhoidal symptoms (Riethmuller et al., 2016). This effect may be probably due to its bioactive compounds. In a study by Amaral et al. (2005) in which the effect of hazelnut tree leaves (*Corylus avellana* L.) from different subspecies on its phenolic composition was investigated, eight phenolic compounds were detected in hazelnut tree leaves, as follows; 3-caffeoylquinic acid, 5-caffeoylquinic acid, caffeoyltartaric acid, ρ-coumaroyltartaric acid, myricetin 3-hexoside, myricetin 3-rhamnoside, quercetin 3-hexoside, quercetin 3-rhamnoside, kaempferol 3-rhamnoside and among these phenolics, myricetin 3-rhamnoside (10.60-18.24 g/kg, dry basis) and quercetin 3-rhamnoside (1.57-4.68 g/kg, dry basis) were found to be dominant phenolics (Amaral et al., 2005). The same phenolics, plus 3-caffeoylquinic, and caffeoyltartaric acids were reported in *Corylus avellana* (Fertille Coutard, Daviana, and M. Bollwiller cultivars) by Oliveira et al. (2007). The researchers also examined the antioxidant and antimicrobial activities of hazelnut tree leaves. It was found that they inhibited more than 93.1% of DPPH at 0.5 mg/ml concentration compared to BHA (96% at 3.6 mg/ml) and α-tocopherol (95% at 8.6 mg/ml). It has an antimicrobial effect against *Bacillus cereus*, *Bacillus subtilis*, *Staphylococcus aureus*, *Escherichia coli*, *Pseudomonas aeruginosa*, and *Klebsiella pneumoniae*, but not toward *Candida albicans* and, *Cryptococcus neoformans* (only cultivar M. Bollwiller was effective at 100 ml/mg) (Oliveira et al., 2007). Riethmuller et al. (2013) evaluated the effect of methanol and ethyl acetate solvents on their bioactive compounds the leaves of *C. avellana* L. Among their bioactive compounds, six flavonoid glycosides (myricetin-3-O-hexoside, myricetin-3-O-rhamnoside, quercetin-3-O- hexoside, quercetin-3-O-rhamnoside, kaempferol-3-O-rhamnoside, kaempferol-di(desoxyhexoside), rosmarinic acid, and a caffeoyl-hexoside derivative were characterized in the methanolic extract while five flavonoids (only not myricetin-3-O-hexoside), and rosmarinic acid were identified in the ethyl acetate extract. Myricetin-3-O-rhamnoside was the predominant flavonoid in both solvents (37.7 μg/mg for methanol, 72.64 μg/mg for ethyl acetate). The research also pointed out that the most suitable solvent is ethyl acetate in terms of the amount of flavonoid compounds (Riethmuller et al., 2013).

In a study by Shahidi et al. (2007), in which the extraction process was carried out in a water bath at 80 °C with 80% ethanol, the amounts of gallic acid, caffeic acid, coumaric acid, ferulic acid and sinapic acid in hazelnut leaves (*Corylus avellana* L.) were found to be 157 mg CE (catechin equivalent)/g, 362 mg CE/g, 884 mg CE/g, 237 mg CE/g, and 247 mg CE/g, respectively. It has been found that it might inhibit the oxidation of LDL cholesterol by 61% at 50 ppm while do catechin 53% and can prevent DNA damage by scavenging the hydroxyl radical up to 50 ppm (Shahidi et al., 2007).

Corylus colorna L., known as Turkish hazel, is rich in flavonoids (Benov & Georgiev, 1994). Riethmuller et al. (2014) investigated the antioxidant activity in different parts, including leaves, bark, catkins, and involucre. They found that the total phenolic, tannin, and flavonoid content in the leaves were 0.94 g/100 g, 0.38 g/100 g, 0.49 g/100 g, respectively. In their study, the derivatives of quinic acid, myricetin, quercetin, and kaempferol were identified and characterized in the extracts from the leaves with methanol and ethyl acetate solvent. The research showed that there was a correlation between the phenolic content of the extracts observed in the research and their antiradical activity (Riethmuller et al., 2014).

Another species of *Corylus* is *Corylus maxima*, a type of hazelnut tree common in the Balkans, Southeast Europe, and Southwest Asia. Riethmuller et al. (2015) conducted research to characterize the bioactive compounds in *Corylus maxima* (leaves and bark). The authors found that the total phenolic, tannin, and flavonoid content in the leaves were 2.43 g/100 g, 0.10 g/100 g, 0.96 g/100 g, respectively. Catechin/epicatechin, myricetin-3-O-rhamnoside, myricetin-3-O-hexoside, kaempferol-glucuronide, quercetin-3-O-rhamnoside, kaempferol, kaempferol-3-O-rhamnoside, and kaempferol-(di)desoxyhexoside were determined in the extract of the leaves and bark with methanol and ethyl acetate and found that the major phenolic compounds are quercetin-3-O-rhamnoside and myricetin-3-O-rhamnoside in both solvents. In the study, the amount of 3-O-rhamnoside in the methanolic extract was 30.01 μg/mg while for the ethyl acetate was 16.9 μg/mg and also a higher amount of quercetin-3-O-rhamnoside (19.8 μg/mg) was obtained by the methanolic extract than by the ethyl acetate (8.2 μg/mg) (Riethmuller et al., 2015).

Cerulli et al. (2018) investigated the metabolomic fingerprinting of the leaves of *Corylus avellana* L. (Tonda di Giffoni, Italian cultivars) by applying the technique of maceration, infusion, and SLDE-Naviglio extraction (Naviglio et al., 2019), which is an eco-friendly solid-liquid extraction technique, allowing us to apply different pressure at different times within a short time compared to maceration and infusion. The extracts with the highest phenolic content were evaluated at optimum extraction conditions, which are 50% ethanol, 1:30 (solid:solvent ratio), 10 h for the maceration; 10:100 for the infusion, and 8 min for the SLDE-Naviglio extraction, and it was found that the phenolic content in each extract was 608.10 mg GAE (gallic acid equivalent) /g extract, 170.57 mg GAE/g extract, 471.80 mg GAE/g extract, respectively. Although the highest phenolic content was in the maceration process, the highest radical scavenging activity (100.33 μg/mL EC_{50} for DPPH, 1.62 mg/mL for TEAC value) was observed in the extract with SLDE-Naviglio extractor as compared to the maceration, the infusion, and quercetin 3-O-rhamnopyranoside. The amount in which it did not show the cytotoxic effect, at 5 μg/mL, 42% inhibition was achieved on pyocyanin, which triggers reactive oxygen species and inflammation.

In addition to flavonoid glycosides (kaempferol 3-O-rhamnopyranoside, quercetin 3-O-rhamnopyranoside, myricetin 3-O-rhamnpyranoside, kaempferol 3-O-rhamnopyranoside), Giffonin A-T, known as diarylheptanoids, were detected in the leaves (Cerulli et al., 2018). Diarylheptanoids, secondary metabolites in plants, are another bioactive compounds contributing to the antioxidant activity of hazelnut tree leaves. They are composed of a 1,7-diphenylheptane skeleton and can be classified into subgroups; linear and macrocyclic, depending on their chemical structure (Figure 2) (Vanucci-Bacqué & Bedos-Belval, 2021). They have been proven to exhibit many biological activities such as anti-inflammatory, anti-carcinogenic, anti-oxidant, anti-microbial (Ganapathy et al., 2019).

Diarylheptanoids were identified and characterized in the leaves of *Corylus avellana*, *Corylus maxima*, *Corylus colorna* (Cerulli et al., 2018; Masullo et al., 2015a; Masullo et al., 2015b; Riethmuller et al., 2013; Riethmuller et al., 2014; Riethmuller et al., 2015). Riethmuller et al. (2013) compared the amount of hirsutenone observed in the extract from the leaves of *Corylus avellana* L. with methanol (0.33 μg/mg extract) and ethyl acetate (2.08 μg/mg), and their results indicated that the most suitable solvent to obtain a high amount of hirsutenone (for other diarylheptanoids detected in the study) was ethyl acetate (Riethmuller et al., 2013). The same result for

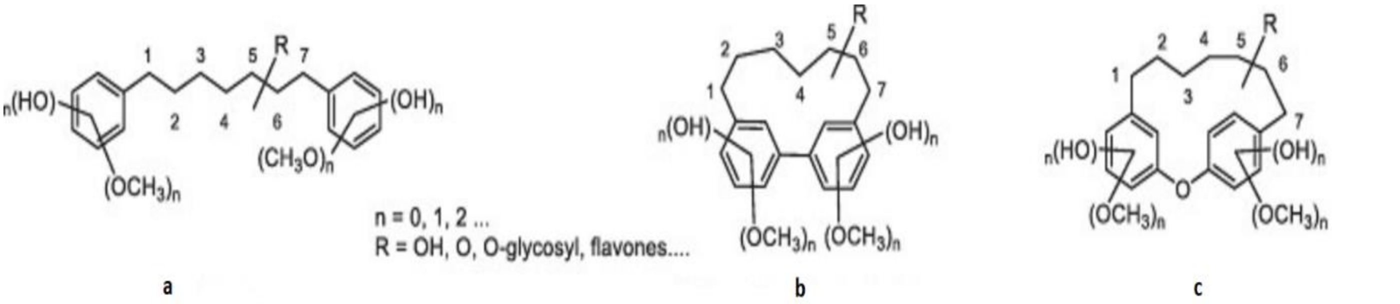

Figure 2. Chemical structure of diarylheptanoids; a: linear skeleton; b, c: macrocyclic skeleton (Vanucci-Bacqué & Bedos-Belval, 2021)

the leaves of *Corylus maxima* was reported by Riethmuller et al. (2015), that is, the content of oregonin (3.40 μg/mg) and hirsutenone (8.39 μg/mg) obtained the extract with ethyl acetate were higher than the methanolic extract (2.21 μg/mg, 0.59 μg/mg, respectively) (Riethmuller et al., 2015).

Giffonin A-I, Giffonin J-P, and Giffonin W-X were isolated and characterized from the leaves of *Corylus avellana* (Tonda di Giffoni, Italian cultivars) and methanolic extract of which tested on plasma lipid peroxidation caused by both H_2O_2 and H_2O_2/Fe^{+2} as well as against oxidation of thiol groups in plasma proteins and human cancer cells (U2Os and SAOs cells). With no cytotoxic on the cancer cells, all the extracts (except for oregonin, and giffonin V only in H_2O_2) exhibited an inhibitory effect on lipid oxidation (H_2O_2 and H_2O_2/Fe^{+2}), which ranged between 2.2-64.3%, 4.0-63.2%, respectively. While the lowest inhibition was found in Giffonin B and J in H_2O_2- or H_2O_2/Fe^{+2}-induced lipid oxidation, the highest inhibition was observed from Giffonin D in both assays compared to curcumin, the most studied diarylheptanoid today (Masullo et al., 2015a; Masullo et al., 2015b; Masullo et al., 2021). Moreover, giffonins decreased protein carbonylation induced H_2O_2 and H_2O_2/Fe^{+2} by inhibiting the oxidation of thiol groups in the protein (Masullo et al., 2021).

Taxane refers to a group of diterpenoid skeletal compounds, which was first isolated from yew-*Taxus* species, by far more than five hundred compounds have been characterized based on eleven different taxane skeletal structures (Nižnanský et al., 2022; Wang et al., 2011) (Figure 3). One of the widely studied taxane types today, paclitaxel (Taxol is tradename, an anti-cancer chemotherapy drug) is used for treating many types of cancer such as breast, lung, prostate, and ovarian. With obtaining in very low amounts from natural sources, it is also possible to produce paclitaxel by semi-synthetic or cell culture methods (Gallego et al., 2017). Paclitaxel, 10-deacetylbaccatin III, baccatin III, 10-deacetyl-7-xylosylcephalomannine, 10-deacetyl-7-xylosylpaclitaxel, 10-deacetyl-7-xylosylpaclitaxel C, 10-deacetylpaclitaxel, 7-xylosylpaclitaxel, cephalomannine, 10-deacetyl-7-epipaclitaxel, paclitaxel C, 7-epipaclitaxel, and taxinine M have been detected in hazelnut tree leaves (Ottaggio et al., 2008). In the defatted-samples, the amount of paclitaxel, 10-deacetylbaccatin III, and cephalomannine were found to be 0.08-0.74 μg/g, 1.48-7.71 μg/g, 0.01-0.16 μg/g, respectively (Hoffman & Shahidi, 2009). The amount of taxane-derived compounds in the leaves is higher than in its shell (Ottaggio et al., 2008).

Another of the bioactive compounds found in hazelnut leaves is alpha tocopherol, which is a type of vitamin E. The amount of alpha tocopherol in hazelnut leaves can vary between 34.6-237.4 μg/g, the highest amount was found in the Moro variety grown in the Saldinia region, in Italy (Sivakumar & Bacchetta, 2005).

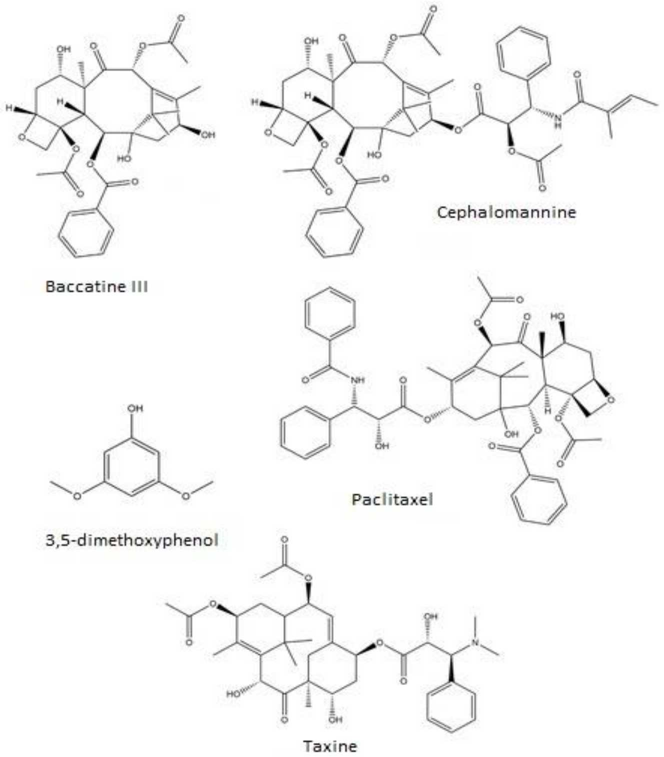

Figure 3. Chemical structure of some taxanes (Nižnanský et al., 2022; Wang et al., 2011)

3. Hazelnut Husk

Hazelnut husk is the plant tissue that surrounds the shelled hazelnut from the outside, is also called as green leafy cover (Alasalvar et al., 2006b), green shell cover (Hoffman & Shahidi, 2009), hazelnut involucre (Rusu et al., 2019), and green outer nut husk (Oguzkan et al., 2018). After the harvest of hazelnut, the hazelnut is separated from the husk by the hazelnut threshing machine in the drying place, and then either may be left in the area where it is dried or burned (Sayar et al., 2019) or used as fertilizer for the hazelnut orchards (Kizilkaya, 2016), plant growing (Özenç, 2006) and used to improve soil quality (Zeytin & Baran, 2003). Furthermore, it can be considered as a raw material, known as biomass, in bioethanol production (Sayar et al., 2019) and may be used as potentially a raw material in value-added biochemical production such as levulinic acid (Sajid et al., 2021), lactic acid (Dusselier et al., 2013), and hydroxymethylfurfural (Van Putten et al., 2013) because of being rich in its lignocellulosic content which is made up of holocellulose (55.1%), α-cellulose (34.5%), and lignin (35.1%) (Çöpür et al., 2007). Monosaccharides such as glucose and, fructose are firstly obtained from lignocellulosic biomass in the presence of acids, alkaline reagents, organic solvents, ionic liquids, deep eutectic solvents, enzymes, and microorganisms and then these sugars are converted into the value-added chemicals (Ashokkumar et al., 2022). Hazelnut husk might be a potentially important raw material source due to its lignocellulosic content, being cheap and widespread availability; it emerged at the rate of one-third of the shelled hazelnut, so it is estimated that approximately 200 thousand tons of it is released annually in Turkey alone (Tufan et al., 2015).

In a study by Alasalvar et al. (2006b), the antiradical and antioxidant activity of hazelnut husk was evaluated, in which 50% ethanol and 50% acetone solvents were used to extract its phenolic acids. According to the results of the study, total phenolic (201 mg CE/g), condensed tannin (542 mg CE/g), total antioxidant activity (1.29 mmol TE/g), and IC_{50} (0.065 mg) obtained from the extract of 50% acetone were higher than those of 50% ethanol extract (156 mg CE/g, 385 mg CE/g, 1.14 mmol TE/g, 0.074 mg, respectively). Moreover, hydrolysis was applied to the hazelnut husk with HCl, and gallic acid was found to be the dominant phenolic acids among free (269 µg/g) and esterified ones (1450 µg/g) (Alasalvar et al., 2006b). Shahidi et al. (2007), determined the antioxidant phytochemicals in by-product of the hazelnut and gallic, caffeic, ρ-coumaric, ferulic, and sinapic acid from the hazelnut husk extract with 80% ethanol were found to be 892 µg/g, 158 µg/g, 1662 µg/g, 327 µg/g, 64 µg/g (both free and esterified) (Shahidi et al., 2007). The highest antiradical activity was observed at 200 ppm for hydrogen peroxide (97%), superoxide radical (99%), and DPPH radical (99.5%). Hydroxyl radical activity, an index of DNA damage, was tested at different concentrations ranging from 5 ppm to 50 ppm, and it was determined that the inhibition increased as the concentration increased, so the highest inhibition against the hydroxyl radical was achieved at 50 ppm (89.9%) (Shahidi et al., 2007). In another study investigating the antimicrobial and antioxidant activity of the hazelnut husk was conducted by Cerulli et al. (2017) and they isolated and characterized the first giffonin T, U as well as giffonin I, citric acid, 1-methylcitrate, trimethylcitrate, kaempferol 3-O-rhamnopyranoside, 3,5-dicaffeoylquinic acid, myricetin 3-O-rhamnopyranoside, and kaempferol 3-O-(4″-trans-pcoumaroyl) rhamnopyranoside from the methanolic extract of the hazelnut husk. The highest and lowest inhibition in the antioxidant activity against lipid peroxidation induced by H_2O_2 and H_2O_2/Fe^{+2} of the isolated compounds was recorded for myricetin 3-O-rhamnopyranoside (44.4%), kaempferol 3-O-rhamnopyranoside (5.7%), and kaempferol 3-O-(4″-trans-pcoumaroyl) rhamnopyranoside (39.7%), myricetin 3-O-rhamnopyranoside (34.1%), respectively. They found that carpinontriol B and giffonin U in the methanolic extract of hazelnut husk were the most effective compounds against *Bacillus cereus*, *Escherichia coli*, *Pseudomonas aeruginosa*, and *Staphylococcus aureus* (Cerulli et al., 2017). Rusu et al. (2019) investigated the bioactive content from the hazelnut involucre, including phenolics and sterolic compounds with D-optimal design. The optimum extraction point for the content of total phenolic, total flavonoid, condensed tannin and antioxidant activity was found to be mixing time 3 min, pH 3 and 50% acetone, as 377.43 mg GAE/g, 43.10 mg QE-quercetin equivalent/g, 280.69 mg CE/g, 1296.51 mg TE/g (TEAC), 292.23 mg TE/g (DPPH), 350.52 mg TE/g (FRAP), respectively. Considering the results of the study, different amounts of individual phenolics and phytosterols were obtained under the different extraction conditions used in the study. The highest amount of epicatechin, catechin, syringic acid, gallic acid, protocatechuic acid, vanillic acid, *p*-coumaric, ferulic acids, hyperoside, quercitrin, and isoquercitrin were determined to be 3.73, 243.02, 5.53, 91.93, 227.37, 25.41, 6.58, 3.97, 51.72, 17.74, and 114.26 µg/g, respectively. In the case of phytosterol, the optimum extraction conditions for stigmasterol (197.31 µg/g), and β-sitosterol (5305.01 µg/g) were stirring time 2, pH 5, and 25% acetone while those for campesterol (45.04 µg/g) was stirring time 3, pH 5, and 25% acetone. Additionally, the extract from hazelnut involucre at the optimum extraction condition was evaluated for enzyme (tyrosinase inhibitory activity and α-glucosidase inhibitory activity) and anticancer activity (A549-human lung adenocarcinoma and T47D-KBluc-human breast cancer). IC_{50} of hazelnut involucre extract for tyrosinase inhibitory activity was 165.17 mg KAE/g whereas those for α-glucosidase inhibitory activity was 0.1 mg/g. Without the cytotoxic effects on human gingival fibroblasts up to very high doses (>300 µg/mL), the hazelnut involucre extract was found to be effective on two cancer cells- A549, T47D-KBluc at 50 and 75 µg/mL (Rusu et al., 2019).

This effect of hazelnut husk on cancer cells may be due to the anticancer compounds it contains. Taxane class compounds such as baccatin III-precursor for the semi-production of paclitaxel have been detected in hazelnut husk as do in the hazelnut leaf. Hoffman & Shahidi, (2009) found the quantities of baccatin III in the ethanolic extract obtained from hazelnut husk of Tombul cultivar, grown in Giresun (in Turkey) after defatting with hexane ranged from 1.10 to 67.77 µg/g (Hoffman & Shahidi, 2009), but not detected the other taxanes containing 10-deacetyl baccatin, 10-deacetyl taxol, cephalomannine, 7-epi-10-deacetyl taxol, and paclitaxel. A similar result was also reported by Oguzkan et al. (2018) and they investigated taxanes in the extract with acetone (at 1:10 g/mL), depending on altitudes and regions in Turkey. They determined that the amount of baccatin III varied from 166.12 µg/kg to 923.64 µg/kg, and the highest quantities were obtained from the extract up to 250 m altitude in Vakfıkebir, in Turkey (Oguzkan et al., 2018).

Cabo et al. (2021) have conducted on the phenolic composition and antioxidant activity of the hazelnut husk of different cultivars (Butler, Grada de Viseu, Lansing and Morell in Portugal) by using different solvents including methanol, water, acetone, hexane, and ethyl acetate. They determined and quantified the phenolics (gallic acid, protocatechuic acid, (−)-epicatechin, quercetin-3-*o*-rutinoside, ellagic acid, luteolin-7-*o*-rutinoside, vanillic acid, kaempferol-3,7-*o*diglucoside, kaempferol-3-*o*-[6-acetylglucoside]-7-*o*glucoside, kaempferol-3-*o*-[6-acetylglucoside]-7-*o*rhamnoside), chlorophyl a-b, and total carotenoids from the hazelnut husk. The total amount of phenolics, flavonoids and total carotenoid obtained with methanol was higher than those with water, acetone, ethyl acetate and hexane whereas the highest chlorophyl content was achieved with acetone. It was emphasized that the hazelnut cultivar and the type of solvent used in the study were effective on the amount of bioactive components from the hazelnut husk (Cabo et al., 2021).

4. Hazelnut Oil Meal

Oil from oilseeds can be obtained using various methods, which can be classified as chemical, high pressure, distillation, and mechanical systems (Ionesu et al., 2016). Hazelnut is an important source of oil raw material and it contains 60% of oil on average, and the main component of its oil is triacylglycerol (Celenk et al., 2020). The triacylglycerol is composed of high levels of oleic acid and, linoleic acid, followed by palmitic and stearic acid (Venkatachalam & Sathe, 2006). In addition to this, it also possesses important food components such as phenolic compounds, B_1, B_2, B_6, β-carotene, lutein/zeaxanthin, tocopherols (α-, β-, γ-, δ-), phytosterols, essential amino acids, serotonin, and minerals (Alasalvar et al., 2009; Alasalvar et al., 2006a; Alasalvar et al., 2003; Celenk et al., 2018; Jiang et al., 2021; Pelvan et al., 2018; Stuetz et al., 2017; Tas et al., 2019; Taş & Gökmen, 2015).

After the hazelnut oil is obtained with a suitable oil extraction technique, the cake remains, which is called as hazelnut oil meal or hazelnut cake in the hazelnut oil industry. The composition of the cake might change on the basis of the extraction methods and solvents used in the extraction process (Geow et al., 2021), in general, the hazelnut cake can include in the range of 81.70-95.87% dry matter, 39.2-54.5% protein, 1-17.38% lipid, 25.20-48.00 % carbohydrate, 4.97-8.64% ash and essential amino acids content account for 17.40-33% of total proteins in the hazelnut cake (Acan et al., 2021; Altop et al., 2019; Aydemir et al., 2014; Gul et al., 2017; Sen & Kahveci, 2020; Xu & Hanna, 2011; Yalçin et al., 2005). Its protein content can be increased up to 94.2% by pretreatments such as acetone washing, alkali extraction and precipitation (Aydemir et al., 2014), or up to 94.8% by proteolytic enzymes (Çağlar et al., 2021b). It has been reported that the essential amino acid content of the hazelnut cake obtained by precipitation after alkaline extraction was up to 37% (Sen & Kahveci, 2020) and arginine was the main dominant essential amino acid, followed by leucine, isoleucine, and phenylalanine (Sen & Kahveci, 2020; Xu & Hanna, 2011).

Because of its high protein content, it might be generally added to diets of animal and fish as a protein source (Karabulut et al., 2019; Kirmizigül & Cufadar, 2019), however, recently, a number of studies have been conducted to determine its technological and bioactivity properties in food technology (Gul et al., 2017; Saricaoglu et al., 2018; Tatar et al., 2015) and to use it an ingredient in food formulations such as hazelnut milk (Gul et al., 2017), functional kefir drink (Atalar, 2019), functional beverage (Sen & Kahveci, 2020), chocolate spread (Acan et al., 2021), chocolate (Bursa et al., 2021), ice cream with the hazelnut milk (Atalar et al., 2021), hazelnut paste (Göksu et al., 2022), and functional yogurt with hazelnut beverage (Gul et al., 2022). What's more it exhibits antioxidant, antiproliferative, antidiabetic, and antihypertensive activity owing to its phenolic compounds and peptides (Aydemir et al., 2014; Eroglu & Aksay, 2017; Simsek, 2021).

Simsek et al. (2017) analyzed the phenolic composition of defatted hazelnut cake obtained from seventeen hazelnut varieties grown in Turkey and found that the total phenolic content of the hazelnut cake varied between 5.29-10.93 mg GAE/g. Mincane had the highest total phenolic content among the seventeen hazelnut varieties whereas the lowest content was found in Foşa. Concerning the phenolic composition in the samples, (+)-catechin was the predominant phenolic, followed by catechol, chlorogenic acid, and quercetin (Simsek et al., 2017). They emphasized that there was a significant difference in terms of phenolic composition and TPC between the hazelnut cakes tested (Simsek et al., 2017). In previous studies, it pointed out that the variety was effective on the phenolic composition of hazelnut skin (Lelli et al., 2021).

A study by Xu & Hanna (2011), in which the extracts from defatted meal in Nebraska was evaluated their physicochemical and bioactivity properties and total phenolic content, tannins, and condensed tannin were found to be 10.7 mg TA (tannic acid)/g, 7.53 mg TA/g, 0.64 mg TA/g, respectively. Furthermore, in the study, it has been stated that it may be an important raw source for minerals, mainly K, P, Ca, and Mg, to human nutrition (Xu & Hanna, 2011).

Bioactive peptides are compounds with bioactivity, which are inactive in the parent protein, which have between 2-20 amino acid sequences with low molecular weight (<6000 kDa) in general (Karami & Akbari-Adergani, 2019). Enzymatic hydrolysis and microbial fermentation are used to produce the bioactive peptides, or they can be synthesized via chemical methods or recombinant DNA technology (Akbarian et al., 2022). The important sources of the bioactive peptides are animals, plants, foods, edible insects, marine organisms, and waste, especially agri-food waste (Chai et al., 2020). Temperature, pH, enzyme used, the length, charge, hydrophobic/hydrophilic properties and type of the amino acid might affect their biological features, such as antioxidants, antimicrobial, antidiabetic, antihypertensive, antiobesity, antithrombotic antiaging, opioid, hypocholesterolemic, and mineral binding activity (Akbarian et al., 2022).

Researchers have reported that hazelnut cake contains bioactive peptides released by various enzymes such as trypsin, pepsin, chymotrypsin, or extracted after pretreatments such as acetone washing or heating (Aydemir et al., 2014; Çağlar et al., 2021b; Simsek, 2021). It has been reported that the combination (86.0%) of acetone washing and heat treatment reduced the protein content of hazelnut meal compared with those of both individual treatment (93.3%, 94.5%) and hazelnut protein isolate (94.2%), respectively, but the antioxidant activity of the combination, measured TEAC and ORAC, was increased. The combination treatment increased ACE (Angiotensin Converting Enzyme) inhibition level by 40% compared to the untreated sample, from 50% to 70%, with IC_{50} 1 mg/mL and, 0.57 mg/mL, respectively (Aydemir et al., 2014).

Depending on the enzyme used in the research, different degrees of free hydrolysis were determined in the hazelnut meal. The degree of hydrolysis for pepsin, papain, thermolysin, bromelain, trypsin, alcalase, chymotrypsin, protamex, trypsin+chymotrypsin (combined) were 74.33%, 60.97%, 50.4%, 46.18%, 26.4%, 23.5%, 21.8%, 18.8%, 13.7%, respectively (Çağlar et al., 2021a; Göksu et al., 2022; Simsek, 2021). Trypsin was used in a study by Gülseren & Çakır (2019), at the end of 4 h incubation with the enzyme, ACE inhibition level of the hazelnut cake increased from 7.6% to around 40% (Gülseren & Çakır, 2019). In a study, pepsin was used to produce bioactive peptides from the hazelnut cake at different times (0, 30, 60 min), its isolate fraction (98.77%) was found to be higher than their hydrolysates when their inhibition levels (98.25%, 97.37%) against ACE were compared, respectively, but the highest IC_{50} (0.22 mg protein/mL) value was observed in the hydrolyzed fraction obtained at 60 min, in comparison to those of the isolate fraction (1.29 mg protein/mL) (Eroglu & Aksay, 2017). Eroglu et al. (2020) found a similar decreasing trend in IC_{50} value, in which the hazelnut protein isolate and its hydrolysates were prepared pepsin and trypsin, IC_{50} of them at 0, 30, 60 and 120 minutes followed the order of 1.47-0.27-0.27-0.26 mg protein/mL; 5.51-0.61-0.56-0.54 mg protein/mL, respectively (Eroglu et al., 2020).

Another study, incorporation of three enzymes (alcalase, protamex, trypsin+chymotrypsin) into a mixture prepared with the hazelnut meal, was performed and their inhibition activity for ACE, DPP-IV (Dipeptidyl peptidase), and α-glucosidase were investigated. It was found that hydrolysates with a 5-20 kDa had higher ACE inhibition value while those with lower than 5 kDa showed higher DPP-IV and α-glucosidase activity (Simsek, 2021). IC_{50} of the peptides with 5 kDa-20 kDa in the study, which were released by alcalase and trypsin+chymotrypsin except for protamex, ranged from 0.10 to 0.18 mg/mL, 0.37 to 1.28 mg/mL, 0-4.76 mg/mL for ACE, DPP-IV, and α-glucosidase, respectively (Simsek, 2021).

Mundi & Aluko (2014) found that <1 kDa to >5 kDa peptide fractions obtained from kidney bean showed better ACE inhibition and antioxidant activity compared to 1-3 kDa, 3-5 kDa fractions, and it was emphasized that this feature may be due to amino acids with hydrophobic and aromatic characters such as valine, isoleucine, leucine, and phenylalanine, tryptophan, respectively (Mundi & Aluko, 2014). Gülseren (2018) obtained twenty-three ribosomal proteins from hazelnut cake, by digesting it with gastrointestinal (trypsin, pepsin, chymotrypsin) and non-gastrointestinal enzymes (lysin, papain and bromelain) and examined their bioactive properties (DPP-IV, ACE) via silico proteolysis using UniProtKB, BIOPEP, and PeptideRanker. According to the results of the research, the bioactivities of peptides produced with non-gastrointestinal enzymes were higher than those of gastrointestinal enzymes, especially papain was the dominant enzyme in comparison of the others in terms of the potential bioactivity, which followed the order DPP-IV (50.68-76.34 %), ACE (16.67-50.23 %), and antioxidative activity (Gülseren, 2018).

Çağlar et al. (2021a) produced the bioactive peptides with ACE inhibitory activity from the hazelnut cake of Tombul hazelnut (Giresun) obtained after the extraction oil by using the enzymatic treatment including trypsin, chymotrypsin, and thermolysin. They determined three peptides by means of LC-Q-TOF/MS and in silico analyses, namely, SPLAGR (trypsin-treated), VPHW (chymotrypsin-treated), and PGHF (thermolysin-treated), which have sixteen, ten, and eleven sites that can bind to the ACE molecule, respectively, compared to VPP (eleven sites binding to ACE) which is one of the most studied dairy-derived tripeptides (Çağlar et al., 2021a). The authors calculated the molecular docking scores for SPLAGR, VPHW, and PGHF through HPEPDOCK (Zhou et al., 2018), which is a web server service used to predict potential interaction between a protein and ligand, for example ACE and the hazelnut protein. According to the results, the molecular docking scores of SPLAGR (−179.023), VPHW (−202.333), and PGHF (− 192.080) were higher VPP (−96.288), indicating that these hydrolysates from the hazelnut cake could be potential inhibitory against ACE (Çağlar et al., 2021a). The research revealed that this interaction was due to amino acids with nonpolar and aromatic rings (tryptophan and phenylalanine) and amino acids with basic and polar character (arginine) (Çağlar et al., 2021a). In another study, molecular docking scores were also found to be higher for LEPTNRIEA (−163.872) and IQVNKENKEFK (−188.424), from hazelnut cake, than VPP (−97.004) based on PeptideRanker scores (Göksu et al., 2022). Additionally, the hydrolysates from the hazelnut cake, which were the fraction of bromelain, papain, and pepsin, were incorporated into the hazelnut paste formulation at ratio of %1, and it was observed that the fractions were not adversely affected by the processes in the production of hazelnut paste and thus maintained their ACE inhibition level (94.21-94.82%) after the production compared to captopril (95.26%) (Göksu et al., 2022).

Çağlar et al. (2021b) determined 256 hazelnut peptides from Tombul hazelnut in Giresun by using proteases (trypsin, pepsin, chymotrypsin, papain, thermolysin and bromelain) through LC-Q-TOF/MS, 7 of which posed a potential high anti-DPP-IV activity according to the results of the silico analyses. They suggested that the interaction between potential bioactivity peptides from the hazelnut cake, namely, PGHF, FMRWRDRFL, APGHF, FFFPGPNK, LSVPNLYVWLCMFY, NSMVGNMIFWFFFCILGQPMCVLLYYHDLMNR, LILVSFSLCLLVLFNGCLG, and DPP-IV could be due to the hydrophobic properties of the amino acids in the hazelnut cake (Çağlar et al., 2021b). Liu et al. (2018) emphasized that leucine is an important amino acid in the inhibition of ACE by hazelnut, as it is a common amino acid in all three bioactive biopeptides, called as AVKVL, TLVGR, and YLVR, produced using alcalase from hazelnut. Moreover, they suggested that cation–π interaction which is a non-covalent bond, as well as electrostatic force and hydrogen bonding, play a role in the interaction between ACE and these three (AVKVL, TLVGR, YLVR) hazelnut hydrolysates (Liu et al., 2018).

The plastein reaction is a chemical reaction whereby peptide bonds in proteins or peptides are hydrolyzed with proteases such as alcalase, papain, and bromelain with a high ratio (%20-50, hydrolyzed product:enzyme), and then ultimately it results in a mixture rich in peptides with high molecular weight through the formation of new peptide bonds between partially hydrolyzed peptide chains, which is known as plastein. Structural and biological changes can occur in plastein at the end of the reaction, which include its particle size, surface hydrophobicity, antioxidant, and ACE inhibition activity and so on (Udenigwe & Rajendran, 2016).

It has been reported that the plastein reaction is a suitable method for modifying hazelnut proteins. Song et al. (2023) modified the hydrolysates from the hazelnut with the plastein reaction by using alcalase in their study, and they found that the ACE inhibitory activity of the plastein (60.74%) was higher than hydrolysates of the hazelnut (41.43%). Moreover, they found that the ACE inhibitory activity of the mixture obtained after the reaction (93.56%) was much higher than YLVR (52.58%) which was used as a substrate to perform the plastein reaction with alcalase (Song et al., 2023).

In addition to having their ACE inhibitory activity, peptides from the hazelnut can also exhibit antioxidant activity, especially those containing methionine, tyrosine, and tryptophan residues at the located C-terminal (Shi et al., 2022). Along with emphasizing the importance of amino acid composition and sequence in a peptide, Shi et al stated that in peptides obtained from the hazelnut hydrolysates, the C-terminus and order of the tyrosine amino acid can affect the functional properties of the peptide (Shi et al., 2022).

5. Conclusion

Hazelnut, which has an important commercial value around the world, is processed into various products. During the process, by-products such as hazelnut tree leaf, husk, skin, and oil meal emerge, which possess the bioactive compounds such as phenolics, dietary fibre, bioactive peptides, and fatty acids. These substances have been proven to have the antioxidant, antiradical, antimicrobial, and antihypertensive effects in both vitro and silico. Therefore, bioactive compounds to be obtained using environmentally friendly extraction methods and solvents, such as ultrasound extraction or deep eutectic solvent, which are the basis of the circular economy, can be used in the food industry, pharmacy, and cosmetics industry. More research should be conducted to test their stability in different medium including food matrix or emulsion or exposed to different heating and its time.

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