POTENTIAL USE OF PLANT-BASED BY-PRODUCTS AND WASTE TO IMPROVE THE QUALITY OF GLUTEN-FREE FOODS

Running title: The exploitation of by-products to improve gluten-free foods

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ABSTRACT

Food industries generate a high amount of waste and by-products, whose disposal has a negative impact on both the environment and the economy. Plant-based waste and by-products are rich in bioactive compounds such as dietary fibre, proteins, essential fatty acids, antioxidant compounds, vitamin and minerals, which can be exploited to reduce the nutritional deficiencies of gluten-free products. The latter are known to be rich in fats, carbohydrates and poor of bioactive compounds; in addition, the absence of gluten affects negatively textural and sensory properties. Several attempts have been made so far to improve the quality of gluten-free products using alternative flours, additives or by adopting innovative technologies. The possibility of exploiting plant-based by-products would represent a possibility to improve both the nutritional profile and the overall quality of gluten-free foods by further enhancing the sustainability of agri-food system. After examining in detail the composition of plant-based by-products and waste, the objective of this review was to provide an overview of the effects of their inclusion on the quality of gluten-free products (bread, pasta, cake/muffins, biscuits and snacks). The advantages and drawbacks regarding the physical, sensory, and nutritional properties were critically evaluated.

Keywords: gluten-free, by-products, waste, functional compounds, dietary fibre.

INTRODUCTION

The past years have been marked by an increase in the incidence of coeliac disease which has led to an increment of gluten-free (GF) products demand. Coeliac disease is an autoinflammatory enteropathy of the small intestine, caused by ingestion of gluten and manifests itself in genetically predisposed subjects (adults and children).[1] Currently the only solution to coeliac disease appears to be a strict adherence to a lifetime GF diet.[2]

Gluten is a tri-dimensional protein complex whose main determinants are prolamins and glutenin proteins present in some cereal flours. Prolamins are named respectively gliadin in wheat, secalin in rye, hordein in barley and avenins in oats and they are responsible of immune reaction in coeliac patients.[3] Only after moisturizing and kneading the flours with water, the technological gluten properties appear, giving viscosity, cohesion and elasticity to the dough and influencing the structural, sensory and nutritional properties of the final products.[4]

The GF products currently on the market are mainly made of rice, corn, sorghum, teff and pseudo-cereal flours and/or starches, with the addition of additives such as hydrocolloids, emulsifiers, proteins and enzymes to mimic the textural properties given by gluten.[5] Several studies highlight the nutritional deficiencies that characterize GF products.[6–8] Coeliac patients suffer from important deficiencies in dietary fibre, B12 and D vitamins, calcium, iron, zinc and magnesium, coupled with an excessive intake of simple carbohydrates and fat, including saturated ones.[9,10] The imbalance in macro and micronutrients exposes patients to a higher risk of non-communicable diseases such as obesity, diabetes, and cardiovascular diseases.[11] In addition to nutritional deficiencies, GF products are characterized by poor structural and sensory properties. Usually, GF foods have an unpleasant texture (low volume, high hardness, crumbling texture), pale colour, due to the conventional ingredients used, and a poor mouthfeel and flavour. [12,13,14]

At the same time, the circular economy is becoming an increasingly global trend. The aim of the circular economy is to switch from a linear production system to one in which waste and by-products are reused, based on the equation "waste=food".[15] Agricultural waste and by-products refers to plant or animal residues that are not (or not further processed into) food or feed and whose management may represent, if not correctly executed, an environmental and economic problem. [16,17]

Processing fruit, vegetables, cereals, oilseeds, and legumes, results in numerous by-products including: peel, leaves, seeds, bran, kernel, pomace, and oil cake[18], which could provide antioxidants, vitamins, minerals, along with dietary fibre (soluble and insoluble), essential fatty acids, bioactive oligosaccharides and oligopeptides.[19,20] Through the use of appropriate technologies, these by-products can be effectively exploited and transformed into innovative ingredients and added to foods, including GF products, with the aim of improving their nutritional profile, evaluating their effects on structural and sensory properties.[21,22] As represented in Figure 1, the reuse of these by-products closes the circle and creates new synergies between different processing industries in the food sector.[19] The rising number of articles published in the last years demonstrates that the exploitation of the plant-based by-products in GF foods is a growing trend and that the GF products improvement is still a challenge. However, no reviews have carried out a comprehensive analysis of all the qualitative characteristics influenced by the addition of these innovative ingredients. In this manuscript, the databases Google Scholar, Scopus, and Web of Science were used to look for search for the relevant articles by using a search strategy that combined the terms "waste", "by-products", "gluten-free", "addition in foods". Therefore, the aim of this review was to provide an overview of current and potential progress made in improving the quality of this category of foods through the reuse of waste and by-products resulting from the processing steps of plant edible matrices.

COMPOSITION OF PLANT-BASED BY-PRODUCTS AND WASTE

A wide range of plant-based food waste and by-products are generated by processing fruit, vegetables, cereals, oilseeds, and legumes, whose characteristics vary according to their specific origin, as reported in the following sections. Table 1 reports the main bioactive molecules extractable from plant-based waste and by-products.

Fruits

The processing of fruit, to produce jams, juices, snacks and salads, produces a high amount of by-products which include peels, seeds, pomace, and bagasse.[23] Despite the high amount of phytochemicals and essential nutrients, these by-products are not usually further exploited in food production.[24,25]

Banana is a tropical fruit largely consumed once it has reached the right size and stage of ripeness. Banana's by-products (green-banana and peels) are rich in bioactive compounds such as flavonoid, vitamins, carotenoids and dietary fibre, this latter is the most abundant. Dietary fibre consists of a soluble (pectin) and insoluble component (cellulose, lignin and hemicellulose); its content and composition vary depending on the stage of fruit ripeness.[26,27] In banana's peel, dietary fibre content can be up to 53% (dry basis percentage), while in the pulp it has been found a maximum value of 15% for unripe bananas and 18% for those in an advanced state of maturation.[27]

An interesting compound mainly present in green banana is resistant starch, that can reach values of 74% in the composition of green banana flour.[28] Resistant starch is mainly composed of amylose, it reaches the colon intact and is fermented by probiotic bacteria producing short chain fatty acids, mainly butyrate, known to have a preventive effect against the colorectal cancer.[29] Moreover, from a technological point of view, banana peel dietary fibre concentrate is characterized by a high water holding capacity (WHC) and oil holding capacity (OHC), making it a potential food additive, capable of changing the viscosity, texture or to increase the oil retention and cholesterol absorption of foods in which it is added.[26] Also passion fruit peel, which represents 60% of the entire fruit, has good technological properties due to its content of pectin (11-20%)[30], and it can be used to replace some commercial hydrocolloids.[31]

Apples and oranges are mainly used for the production of juices, generating a large amount of pomace. Pomace is what remains after juicing: apple pomace includes peel, pulp and seeds, while orange pomace, contains peel and pulp.[32] Even these by-products are characterized by a high amount of total dietary fibre: Kırbaş et al.[33] found a value equal to 82.2% on dry matter (d.m.) for orange pomace (cv. Washington navel) and 64.84% d.m. for apple pomace (cv. Red Delicious), making them suitable to be used as innovative ingredients to improve the nutritional and technological properties of food products. Orange pomace shows a higher protein (9.81% d.m.) and ash content (4.29% d.m.) than apple pomace, which is characterized by 64.84% d.m. of dietary fibre, 3.57% d.m. of protein and 4.29% d.m. of ash.[33]

Furthermore, agriculture by-products are a valuable source of natural antioxidants. It has been highlighted a variable content of polyphenols in apple pomace (66.2 to 211.9 mg of total phenols/100 g), which consists primarily of catechins and proanthocyanidins followed by hydroxycinnamates, flavonols, dihydrochalcones and anthocyanins and is influenced by the method of juice production.[34,35] It was noted that polyphenols concentration was higher in the pomace from the production of clear juice by the action of pectinases, used for clarifying the juice, that break the link between polyphenols with pectin or other cell wall structures, increasing extractable polyphenols.[34] The recent increase in the consumption of coconut-derived products, such as milk or oil, has led to an increase of waste from their processing. However, the high content of fats, dietary fibre, protein and minerals such as calcium, sodium, potassium and magnesium, in coconut waste makes it suitable as a food ingredient.[36]

Pomegranate is a very ancient fruit belonging to the *Punicacea* family, consumed fresh or used for the production of juices.[37] Seeds are the main waste resulting from pomegranate processing, and their reuse as food additives is becoming increasingly widespread.[38,39] The growing interest in the reuse of pomegranate seeds derives from the fact that they have an interesting lipid composition, predominantly polyunsaturated, as found by Bustamante et al. [40] The most represented fatty acid was punicic acid (65.1-78.4%), followed by linoleic acid (5.1–8.1%), oleic acid (5.2-6.8%) and palmitic (2.3-4.0%).[40] Punicic acid is a conjugated α-linoleic acid and several beneficial properties were attributed to it for human health. [41] Specifically, punicic acid has proven to have antioxidant, anti-inflammatory, anti-obesity and chemo-preventive properties. [42] In addition, pomegranate seeds are characterized by a high levels of antioxidants such as ellagic acid, 3,3’-di-*O*-methylellagic acid, chinic acid, and malic acid. The other compounds found in pomegranate seed were *p*-hydroxybenzoic acid, methyl 2-[cyclohex-2-en-1-yl (hydroxy) methyl]-3-hydroxy-4-(2-hydroxyethyl)-3-methyl-5-oxoprolinate, and 3-oxoocta decanoic acid.[43] Olive by-products and waste (olive pomace, olive mill wastewater and olive leaves) represent another important source of antioxidants, specifically phenolic compounds.[44] According to Difonzo et al.[45] the main phenolic compounds of olive leaves (*Olea europea,* cv. Coratina) are oleuropein and hydroxytirosol glucoside. Likewise, olive mill wastewater is particularly rich in phenolic compounds, with hydroxytyrosol (66.5%) as the major compound. Minor concentrations of tyrosol, caffeic acid, p-coumaric acid and ferulic were detected by Paulo et al. [46] Also grape by-products, i.e. skin and seeds, are a source of phenolic compounds and fibres (50-

75% d.m.).[47] However, the composition and content in bioactive compounds can be influenced by various factors including: genotype, growth conditions, degree of ripeness of the fruit.[48] Grape peel is a source of anthocyanins, hydroxycinnamic and hydroxibenzoic acids, cathechins and flavonoids (flavonols, anthocyanins and flavan-3-ols) tannins, proanthocyanidins and stilbens. In particular, anthocyanins, hydroxycinnamic acids, catechins and flavonols displayed a high antioxidant activity, contributing to the inhibition of low-density lipoprotein oxidation. [49] The ability of grape polyphenols to reduce starch digestion, lowering the glycaemic index of food they are added to, has been also demonstrated.[50] Grape peel contains also cellulose, hemicelluloses, pectin, sugar, proteins and minerals (potassium, magnesium and calcium), which could be exploited for nutritional, functional and structural improvement of foods.[49]

Berry pomace and seeds, e.g. chokeberry, black currant, raspberry, strawberry and blueberry, as well as grape by-products present a high amount of fibre and polyphenols, especially anthocyanins, and a good level of proteins (∽12% d.m) and fat.[51,52]

Also coffee by-products, such as pulp, husk and silverskin, the latter generated during the roasting phase, can be considered a sustainable source of macro and micronutrients. Even for these by-products one of the most abundant macronutrient is dietary fibre, which reaches the highest value in silverskin (62.4%) as reported by Janissen et al.[53] However, they also contain a good amount of proteins, minerals and bioactive compounds (tannins, caffeine, chlorogenic acids and melanoidins formed by Maillard reaction during the roasting process).[54]

Vegetables

The production of processed vegetables generates a high amount of waste and by-products, sources of nutrients and bioactive compounds.

Carrot juice processing generates 30-50% of pomace,[55] mainly consisting of fibres that can reach values of 64.15% d.m. Insoluble fibre represents the largest fraction, accounting for 79% of the total dietary fibre, the remaining part being soluble fibre.[56] Other bioactive compounds that can be found in carrot pomace are phenolic compounds, carotenoids, uronic acids and neutral sugars.[55] Carotenoid are natural pigments, with antioxidant properties and known to be precursors of vitamin A.[57,58] The content of these compounds is affected by the conditions of the drying process needed to make carrot pomace microbiologically stable. Ortega et al.[56] found a carotenoid content of 92.64% d.m. in fresh pomace, on the other hand values of 74.10% and 65.74% were found in carrot pomace dried by microwaves and hot air, respectively. Also the phenolic compounds are thermolabile, resulting in a lower concentration in dried carrot pomace than the fresh one; the reduction of these two bioactive compounds would justify the lower antioxidant activity found in dried pomace.[43]

Among the vegetables, broccoli by-products have recently gained interest because of the presence of numerous bioactive compounds common to the majority of vegetables by-products, but in particular for the presence of glucosinolates (GLS).[59] Glucosinolates are a class of sulphur-containing glycosides that are found in a limited number of plant families, with several and relevant beneficial effects for human health.[60] These compounds have to be hydrolysed to isothiocyanate by the myrosinase enzyme to become biologically active.[61] Broccoli leaves powder (*Brassica oleracea L.*, cv. italica) shows a surprisingly high content in proteins (28.99%) and minerals (10.65%) and 5 μmol g^{-1} d.m. of total GLS. The HPLC analysis revealed the presence of 9 GLS, with a predominance of glucobrassicin (indole GLS) and glucoraphanin (aliphatic GLS), which represent 30% and 25% of the total GLS, respectively.[61]

According to FAOSTAT data[62], potato world's production reached values of around 370 millions of tons in the year 2019. Its processing, to produce snacks, chips, starch products or frozen products, leads to a high amount of by-products, mostly peels. Several studies have demonstrated the richness of potato peel in phenolic compounds;[63,65] from the analysis performed by Singh et al. [66] on potato peel extract emerges a prevalence of caffeic acid, followed by gallic acid, protocatechuic acid and chlorogenic acid.[66] In addition, the oxygen radical absorbance capacity (ORAC) test carried out by Joly et al.[67] to estimate the antioxidant activity of the various components of potato, showed a high value for freeze-dried potato peel extract, without significant differences for the solvent used, emphasizing the potential use as natural antioxidants. Also tiger nut waste, which derives mainly from the production of Horchata de chufa (traditional Spanish milk-like beverage) has shown interesting properties due to the high richness in dietary fibre (99.8% IDF) which has exceptional water and oil retention capacities with emulsifying capacity and stability. These characteristics make tiger nut co-products a suitable ingredient for the formulation of dietetic food and ensure emulsion formation and stability during prolonged storage.[68]

Cereals, oilseeds and legumes

Rice (*Oryza sativa*) is a cereal, whose production is widespread all over the world and it is commonly endured to refining processes from which by-products, such as rice bran and rice fragments, are derived. Rice bran is a good source of dietary fibre (27%), unsaturated fat (17%), protein (14.6%), minerals (7%) and vitamins (thiamine, riboflavin and niacin). Regarding minerals, they are concentrated mostly in the outer layers, with phosphorus as the major one.[69]

Several studies on rice bran highlighted its beneficial health effects, in particular: hypocolesterolemic effects due to the presence of unsaponifiable like phytosterols, oryzanols and tocotrienols; preventive effect against coronary heart disease, and colorectal cancer due to dietary fibre and phytosterols, while oryzanol acts as protection against skin damage.[70] Fibres extracted from defatted rice bran have proven to possess functional properties such as water and fat binding capacity. In particular, defatted rice bran fibres have a higher fat-binding capacity than FIBREX (commercial fibre preparation). [71]

Chia seed cake and hemp seed cake are two by-products resulting from oilseed extraction. They are valuable sources of proteins, total dietary fibre and polyphenolic compounds.[72,73] Regarding chia meal, even if total dietary fibre is largely composed of insoluble fibre with a low amount of soluble fibre, it has a high water holding capacity due to the mucilage that acts like soluble fibres.[74]

Hemp seed cake, beyond a high amount of fibre, is characterized by a relevant content of proteins: albumins and edestin, rich in essential amino acids. Edestin has a high biological value due to its structure similar to serum globulin. Therefore, both these by-products have the potential to be used for the production of functional foods.[75]

Legumes by-products result from industrial processes and are known to be an excellent source of proteins and dietary fibre.[76] Okara is a by-product from soya milk extraction or tofu production, with a yellow-white colour and a delicate taste. Its composition reveals a high percentage of total dietary fibre (58 g 100 g^{-1} d.m.), mainly insoluble (55.63 g 100 g^{-1} d.m.), a considerable percentage of proteins (15.31 g 100 g^{-1} d.m.) and a low content of crude fat (5.90 g 100 g^{-1} d.m.) as reported by Lu et al.[77]. According to Lu et al. [77] the addition of okara powder to food, e.g. noodle, steamed bread and white bread, determines an hypoglycaemic effect due to dietary fibre, preventing diseases like type 2 diabetes, coronary heart disease or some cancers. Also carob by-products such as germ and seed peel are rich in dietary fibre and protein but, overall, they are an important source of phenolic compounds. Rico et al.[78] have found respectively 47.06 μmol gallic acid equivalents (GAE) g^{-1} d.m. in carob's germ and 99.72 μmol GAE g^{-1} d.m. in carob seed peel, which confers a high antioxidant activity, anti-inflammatory capacity, and anti-hypertensive activity. Even by-products deriving from legume milling, such as chickpea hull, contain high amounts of dietary fibre (65-82 g 100 g^{-1}) and antioxidants such as phenolic compounds and carotenoids. Chickpea hull from the subtype Apulian black contains also relevant levels of anthocyanins (225 mg kg^{-1} cyanidin 3-*O*-glucoside d.m.) making them suitable to be exploited by food industry[79]

WASTE AND BY-PRODUCTS AS INNOVATIVE INGREDIENTS OF GLUTEN-FREE FOODS

As already mentioned, GF products are in many instances characterized by deficiencies in several aspects: structural, sensory and nutritional. Therefore, with a perspective of an improvement of GF products and an increment of food industries sustainability, some studies have used plant-based waste and by-products as innovative ingredients to achieve different purposes.

Effect on physical and sensory properties of GF foods

The absence of gluten significantly affects the structure and sensory properties of GF products.[12] According to Grigor et al.[80] the addition of fibre could improve or worsen physical and sensory properties of cereal foods, based on the initial acceptability of the product. Table 2 summarizes the main effects of adding waste and plant-based by-products to GF foods.

Bread

The addition of fibre-rich ingredients could cause a change in rheological, structural and consequently sensory properties of GF bread due to their interaction with proteins, starch and water.[81] Structural properties of bakery products are strongly influenced by rheological properties of batter. Key values about batter are provided by oscillatory test, such as elastic modulus (G'), which refers to the material response as a solid and viscous modulus (G''), which refers to the material response as a fluid.[82] The addition of 5.5% of orange pomace leads to high G' modulus with a consequent higher batter consistency, firmness and elasticity than the control and lower gelatinisation rate.[83] In a subsequent study, O'Shea et al.[84] focused on the influence of orange pomace on the structural properties of GF bread. The development of an experimental design made possible to evaluate the influence of three variables: percentage of orange pomace added, quantity of water added and proofing time. A low inclusion of orange pomace (2-4%) led to better textural properties of GF bread. Specifically, from the experimental design developed, it has emerged that the combination of 2% of orange pomace, 88.75% of water inclusion and 42.5 min of proofing time, resulted in a bread with a significantly lower hardness than the control, made of rice flour and potato starch, and a consequently softer texture. Authors explanation refers to the water binding capacity of orange pomace fibres, which leads to a low loss of moisture, delaying the degradation of starch. However, the maximization of orange pomace inclusion, made it necessary a later phase of optimization, from which it has assessed that combining 5.5% of orange pomace, 94.6% water and 49 min of proofing, allowed to obtain a good loaf volume and acceptable texture. The addition of water is a crucial factor to moderate the effects on the physical properties caused by the addition of pomace.[82]

Rocha Parra et al.[83] found that the addition of different amounts of apple pomace in combination with different levels of water in batter preparation have a strong influence on the rheological properties in terms of viscoelastic behaviour. It was noticed that an increasing level of apple pomace and an inadequate level of water caused a reduction of bread volume, cohesion and resilience probably due to the large capacity of fibres to absorb water. Also Sarawong et al.[85] highlighted the importance of water addition on physical properties of GF bread. However, it was noticed that by keeping the amount of added by-product constant, high quantities of water, result in a decreasing volume and crumb firmness and a crumb pore structure with high number of small and uniform pore. Experimental trials show that the addition of 160% of water with a baking temperature of 180 °C for 90 min were the best conditions for preparing GF bread with 30% of green plantain powder.

Genevois et al.[86] added different quantities of rice bran to a GF bread formulation according to an experimental design, considering three variables (rice flour, coarse and bran). From the results, the bread with 45% of rice flour, 35% and 20% of rice bran showed a higher specific volume and colour, and a lower value of firmness than the other formulations. A key role is played by fibres, which thanks to their nature and interaction with rice flour protein improved volume, firmness, crust and crumb colour of GF bread. Also the addition of tiger nut milk by-products have influenced the batter consistency and the texture of bread. Consequently to the use of this by-product to substitute soya flour in a GF bread formulation, Aguilar et al.[87] observed an increase in G' modulus, producing a consistent batter that, in accordance with the studies previously mentioned, should lead to obtain a bread with higher specific volume. However, the leavening was negatively affected probably due to the excessive consistency of the batter, causing a limited increase in volume and a hard crumb. Therefore only 1.69% of consumers involved in sensory analysis preferred the bread with tiger nut milk by-product.

Cakes and muffins

Cakes and muffins are mainly consumed for breakfast or as a snack during the day. Their quality is expressed in terms of both physical (weight, height, volume, crust and crumb colour, symmetry, crumb structure and texture) and sensory properties.[88]

Majzoobi et al.[89] combined different levels and particle size of carrot pomace to improve the quality of GF sponge cake. The addition of carrot pomace led to an increase in batter properties (viscosity, density, firmness and consistency), which positively affected texture, reducing hardness, cohesiveness, gumminess and increasing the chewiness of the cakes. Likewise, also the sensory characteristics were improved. The addition of carrot pomace powder resulted in a larger appreciation of crust and crumb texture by the panellists. Moreover, the enhancement of Maillard and caramelisation reactions, as a result of increased sugar and protein content, resulted in improved flavour of final products.

The eggless GF rice muffins enriched with black carrot pomace (6%) and xanthan gum, produced by Singh et al.[90], resulted in lower firmness, cohesiveness, springiness and gumminess than the control. Likewise, GF cakes based on apple pomace using optimized quantities of starch, xanthan gum and sugar, had lower hardness, springiness, cohesiveness and gumminess than rice cakes but higher hardness if compared with wheat cake.[91] However, sensory analysis displayed that the addition of apple pomace, gained the highest taste score in the GF cakes due to the flavour and aroma of the fruit. The comparison between GF muffins added with commercial orange fibre (COM) and orange fibre obtained from orange peels, using a combined drying system: hot air (HAD) and microwave (MW) showed no significant differences in some textural parameters (cohesiveness, resilience and springiness) with the exception of hardness and chewiness, which resulted higher in HAD+MW muffins than COM. This result could be explained by the higher holding water ability of HAD+MW fibre than COM. On the contrary, sensory analysis revealed a greater acceptability of HAD+MW muffins than COM ones, for its attractive colour, appearance, flavour and chewiness.[92] Moreover, it was observed that the progressive addition of green banana flour (GBF) by Türker et al.[93] and extruded broken bean flour by Gomes et al.[94] to produce GF cakes, resulted in a dose-dependent reduction of volume and colour parameter $L*$, while increased density. However, while the progressive addition of increasing amounts of extruded bean flour did not significantly affect sensory properties, GF cakes with an inclusion of 15% and 20% of GBF received lower scores from hedonic tests than GF cakes with 5% and 10% of GBF added.

Finally, powders of grape and pomegranate seeds were added to GF cakes.[95] The addition of the two seeds powders caused a decrease of volume, symmetry, uniformity index and an increase of hardness of the cakes compared to the control (rice based), especially for the cake with addition of pomegranate seeds powder. Also the colour of crumb and crust was affected by the addition of the latter powder, decreasing $L*$ and $b*$ parameters, and increasing $a*$. From a sensory point of view, there were no significant differences between the cakes supplemented with grape and pomegranate seeds powders and the GF control cake in terms of appearance, texture, taste-odour and overall acceptability, except for mouthfeel that was higher in control cake.

Pasta

Pasta is consumed worldwide for its palatability, versatility and long shelf-life. A key role in the determination of pasta quality is played by both raw materials and processing conditions.[96] Parameters that influence the quality of pasta include: solid loss during cooking, stickiness, and firmness. [97] The absence of a gluten network determines a low cooking quality in GF pasta with a subsequent high stickiness and cooking loss, thus the addition of commercial or innovative texturizing ingredients are necessary for improving GF pasta quality. [98]

Physical and sensory properties of GF pasta processed with potato pulp, a by-product obtained by sedimentation from the water used to wash the raw material for chips production, were evaluated by Bastos et al.[99] Seven different formulations were produced with different amounts of dried and extruded potato pulp and amaranth flour. In general, it was observed a tendency to increase solid loss and reduce mass values with increasing inclusion of dried potato pulp. On the contrary, with lower amount of dried potato pulp and higher amount of extruded potato pulp and amaranth flour a lower solid loss values and higher values of mass increment were observed. According to the authors, this result derives from a larger capacity of extruded potato pulp to absorb water than dried potato pulp and higher protein content from amaranth flour that leads to the formation of a network, between protein and starch, sufficiently stable to avoid starch leaching. Moreover, the formulation with 65% of dried potato pulp, 10% of extruded potato pulp and 25% amaranth flour showed a lower cooking time than non-GF spaghetti, a more intense and darker colour and a high sensory acceptance score.

The replacement of 10% and 20% of rice and corn flours with passion fruit peel flours by Ribeiro et al.[100] affected physical and sensory properties of GF spaghetti. The authors found an increase in cooking time and soluble solid loss in comparison with the control (corn and rice flour). It was observed that with an increasing amount of passion fruit peel flour (PPF) the colour parameter $a*$ increased (from -0.24 to 4.05) while luminosity $L*$ decreased (from 79.47 to 58.45) compared with the control. From a sensory point of view, a deterioration was observed for all the parameters considered (taste, flavour, texture, colour and overall appearance), proportionally to the quantities of PPF added. Mirhosseini et al.[101] found similar results regarding colour and sensory properties with the addition of durian seed flour to GF spaghetti. In addition, textural properties were improved, through an increment of uncooked and cooked pasta hardness and adhesiveness decrement. Patiño-Rodríguez et al. [102] evaluated the possibility to use unripe banana flour as innovative ingredient for the production of GF spaghetti. Specifically, they produced two different formulations: one with whole unripe banana (PFP) and the other only with pulp (PF). Both of them were compared with commercial GF spaghetti. Textural analysis of cooked spaghetti displayed a greater hardness of PFP spaghetti and lower chewiness than commercial one. Cohesiveness and elasticity did not differ in the innovative spaghetti, but commercial sample presented the highest values. Through an optimization process of GF pasta, Fradinho et al.[103] produced a GF pasta consisting of 20% of rice gel and a 50:50 gel: flour ratio in which cooking loss, stickiness and adhesiveness were minimised and swelling, water absorption and firmness were maximised. These results have emphasized the suitability of broken rice kernels to be used for food application thanks to their good gelling abilities. Chickpea hull was effectively added to GF pasta (8% w/w) improving fibre content and antioxidant activity, without affecting sensory liking.

Snacks

The GF snacks production is still challenging, due to the issues in reaching some specifics physico-chemical and sensory parameters. In this framework, Alonso et al.[104] produced extruded GF breakfast cereals with broken rice flour, passion fruit peel flour and whey powder, and evaluated the influence of two process parameters, moisture content and temperature, on product quality. The best physical results were obtained combining a moisture content of 15% and a temperature equal to 85 °C during the extrusion process. These parameters provided the best expansion index (above 2.6), and the highest a^* value. As regards the purchase intention, 38.6% of the testers would probably buy these breakfast cereals.

The effects of olive leaf and olive mill wastewater extracts addition with two different concentration of phenols (500-1000 mg kg^{-1}) on the properties of GF breadsticks was studied.[105] The colorimetric and structural parameters were influenced, besides to an increase of phenols content and antioxidant activity. Specifically, the addition resulted in a reduction of a^* for all the samples supplemented with the extracts and a significant reduction of hardness in those added of olive leaf extract, probably due to higher water content than control sample made of rice flour and corn starch.

Effects on nutritional properties of GF foods

The integration of plant-based by-products in GF formulation comes from the increased attention of consumers to their health and the awareness that the latter is strongly influenced by diet. Several products have been the object of researches aimed at nutritional improvement, with baked products (bread, cakes, muffins, biscuits, pasta and extruded-cooked products (snacks, breakfast cereals) as the most studied. Table 3 reports the improvements resulting from the addition of waste and plant-based by-products.

Ostermann-Porcel et al.[106] produced GF biscuits, using okara flour as a substitute of manioca flour. The resultant cookies were characterised by an increased content in proteins, fibre, fat and ash, proportionally to okara flour added. Other GF biscuits using pregelatinized rice flour and 15, 25 or 35% orange pomace blended with 5% of soy protein isolate were produced by Cayres et al.[107] The results showed that the biscuits with 25% and 35% of pregelatinized flours allowed to produce a product that could be claimed as source of fibre according to the Regulation (EC) No.1924/2006[62], because contained more than 3 g of total dietary fibre per 100 g. Several health benefits (prevention of obesity, diabetes, coronary heart disease) are associated to the dietary fibre. However, to ensure these benefits, the American Dietetic Association recommends a daily intake of 25-35 g or an equivalent of 14 g/1000 kcal.[108,109] Moreover, a portion of 30 g biscuits was able to provide the recommended daily intake of some minerals (iron, copper, manganese, phosphorus and sodium). Also the nutritional profile of GF bread obtained by Korus et al.[110] replacing the starches (corn and potato) with defatted strawberry and blackcurrant seed's powders (5-10%) was characterized by a higher content of total dietary fibre than the control. The total dietary fibre was in a range of 4.53-7.41% for bread enriched with defatted blackcurrant seed (5-15%) and in a range of 5.43-8.71% for that enriched with defatted strawberry seed (5-15%) in comparison with the control, which contained 2.99% of total dietary fibre. Moreover, the two supplemented breads showed a higher protein content than the control sample, due to the very high amount of protein in defatted blackcurrant and strawberry seed, respectively 25.3% and 15.7%. As regards polyphenols content, the breads supplemented with defatted seeds showed a significant increase in their polyphenols content and antioxidant activity, respectively 484-1275% and 166-371% higher than control bread.

Likewise, mini sponge cake produced by replacing 2.5, 5 and 7.5% (w/w) of potato and corn starch, in GF control formulation, showed a significant increase in antioxidant activity, in a dose-dependent manner with broccoli leaf powder (BLP) content of each sample. Regarding glucosinolates (GLS) profile found in mini sponge cakes, it was identical to GLS profile of BLP with a major concentration of glucobrassicin and glucoraphanin, 16% and 19% of total GLS in the sample with the highest inclusion of BLP, respectively.[61] In line with the results obtained by Drabińska et al.,[61] also the GF muffins produced by substituting carrot pomace to rice flour base by Olawuyi et al.[111] displayed an enrichment in bioactive compounds (polyphenols and carotenoids), whose content increased in proportion to the amount of carrot pomace added as well as the antioxidant activity.

The addition of green banana flours into layer and sponge cake allowed to obtain products with a better chemical composition without adversely affecting the sensory quality.[28,112]

A lower content in lipids and a higher content in fibre were found in sponge and layer cakes than the control sample. On the contrary, in standard GF pasta it was found a higher fibre content than in green banana pasta, because of the absence of an adequate method to determine the resistant starch, which instead of being considered as dietary fibre, was included in carbohydrate content. Even the GF snacks produced with the addition of cactus peel powder by Miranda et al.[113] had a large fibre content, which was underestimated. Moreover, the snacks with 10% of cactus peel powder inclusion were characterized by a lower amount of fat and higher content of protein than commercial snacks and improved sensory and textural properties.

Fradinho et al.[114] produced a GF pasta with the aim of improving the nutritional performance of the product through the use of potato peel autohydrolysis extract. From the analysis of the raw and cooked pasta emerged that the addition of this extract resulted in an increase in protein and ash content, reducing fat to a level that makes possible to attribute the fat-free claim.[62] Likewise, GF noodles prepared by resistant rice starch with the addition of defatted rice bran (5%) and xanthan gum (2.5%) exhibited the highest protein, fibre and ash content among the GF noodles produced with a low glycaemic index and a high sensory acceptability by panellists.[115]

Other authors[116,117] produced functional crackers with the addition of carob's germ and seed peel and apple pomace, respectively. The addition of these by-products in different concentration in crackers formulation resulted in an increase in fibre and antioxidant activity for both the products, whereas proteins increased only when carob's by-products were added. However, the addition of these by-products caused colour, sensory and textural modifications necessitating to find a compromise between nutritional properties and physico-chemical aspects. Also the substitution of cassava flour with green banana led to a significant nutritional improvement of the crackers.[118] Through the replacement of the cassava flour, in all the formulations occurred a significant increment of ash, total dietary fibre and essential minerals like potassium, magnesium, phosphorus and calcium, proportionally to the green banana flour replaced.

Otherwise, the addition of hemp seed oil press-cake, decaffeinated green tea leaves and chia seed to brown rice flour to produce functional cracker by Radočaj et al.[119] determined an increase in unsaturated fatty acids (mono and polyunsaturated). Nowadays, modern diet is characterized by an imbalance intake of essential fatty acid, with an excess of omega-6 and a low amount of omega-3, at which is associated several health implications, i.e. obesity, cardiovascular disease, cancer and inflammatory and autoimmune diseases. A low ratio of omega-6/omega-3 (2:1) is associated to the prevention of the aforementioned disease.[120,121,122] The hemp seed oil press-cake increased the omega-6 and omega-3 content in all the crackers, obtaining an optimal ratio between 1.66 to 1.83. With regard to the sensory aspect, all the samples were considered acceptable, in terms of appearance, colour and flavour, except for the sample with the addition of 40% of hemp flour and green tea leaves, which received the lowest score (7.8 ± 0.2).

The replacement of 10-50% cassava flour with by-products from virgin coconut oil processing GF flakes resulted in improved nutritional properties. Specifically, the sample with 20% coconut flour and 80% of cassava flour had 8.56 % of fibre, 4.50% of proteins and minerals like potassium, sodium, magnesium and calcium.[36]

Chia seed waste and defatted rice bran from oil extraction were added to GF bread formulation by Zdybel et al.[123] and Phimolsiripol et al.[124], respectively. The addition of 5% of chia seed waste and

10% of defatted rice bran, determined an enhancement of nutritional properties. Defatted rice bran contributed proteins and total dietary fibre (insoluble and soluble) raising their content in the final product. An interesting aspect regards the action of soluble fibres in slowing down the staling kinetic during storage. In fact, the bread supplemented with a commercial rice fibre (Risolubles), which possessed a higher amount of soluble fibres, was characterized by a slower staling. Therefore, the effect of soluble fibres can be exploited to prolong the shelf-life of GF bread. On the other hand, chia seed waste addition increased significantly protein, fat and ash content, while decreasing the energy value if compared with the control. According to the authors, this is a consequence of the high fibre content of raw materials and high levels of moisture in GF breads. Guglielmetti et al.[125] added coffee husk and coffee silverskin extracts into GF breads new formulation, containing inulin and rice protein. The new breads with and without coffee by-products extracts displayed a reduced carbohydrate content, increased total dietary fibre and proteins. The addition of coffee by-product extracts into breads formulation enhanced inhibitory effect on alfa-glucosidase enzyme if compared with commercial GF bread (control) and bread without the addition of extracts, as a result of chlorogenic acid presence in coffee by-product extracts. Results of *in vitro* oral-gastrointestinal digestion indicated a reduction of fermentable sugars (glucose and fructose) in breads added with coffee by-product extracts and a higher antioxidant activity in the innovative breads as consequence of antioxidant peptides, deriving from rice protein, released during digestion process. Meanwhile, the addition of isolated coffee cascara dietary fibre (3- 4.5%) and rice protein (8%) to a GF baking pre-mix allowed to obtain breads that could be labelled as "high in dietary fibre" and "source of protein",[62] with an improved textural and sensory profile.[126]

Littardi et al.[127] produced a coffee parchment-enriched (2%) bread. The obtained bread showed a significant increase in antioxidant activity, six-fold higher than control bread (rice and corn flour), despite the total phenol content did not increase compared to the control. In addition, the results showed a significant reduction of 5-hydroxymethylfurfural in the enriched bread, which is formed in heated food during caramelization and Maillard reaction and to whom are related negative health effects (genotoxic and mutagenic effects on bacteria and human cells).[128] The reduction of this compound can be attributed to the presence of antioxidant compounds. Also the total substitution of wheat flour with partially defatted baru (*Dipteryx alata*) waste flour to produce GF cake led to an improvement of nutritional properties.[129] The cake produced presented a significant increase in protein, fibre and minerals content such as to be labelled as high protein content, rich in fibre and high zinc content. Moreover there was a significant increase in total phenolics, total flavonoids and tannins, which are known to have several health benefits like antioxidant, anticancer, cardioprotective, antidiabetic and obesity through the inhibition of salivary and pancreatic alfaamylase and alfa-glucosidase activity.[130] Similarly, the addition of different percentages of pomegranate seed powder (2.5-10%) determined an increase of phenolic content and antioxidant activity in the GF bread.[131]

CONCLUSION

The focus of the review was to provide an overview of the influence of plant-based waste and by-products on the quality of GF foods (bread, pasta, cakes, biscuits and snacks). Plant by-products have been proven to be a rich and sustainable source of bioactive compounds, which have several health benefits (prevention of coronary disease, obesity, diabetes and cancer). Dietary fibre was found to be the most abundant component in all by-products followed by phenolic compounds. In addition, other bioactive compounds such as carotenoids, glucosinolate, essential minerals, amino acids and fatty acids have been identified on the basis of the by-product nature. Therefore, the integration of these by-products or their extract into GF products has led to a nutritional improvement, mainly by increasing dietary fibre content and antioxidant compounds that are known to be deficient in GF products, and to which is associated a reducing action of glycaemic index of foods, a critical and common aspect of GF products. However, the nutritional improvement, as a consequence of plant by-products addition, is often accompanied by a deterioration of structural and sensory properties of the final products. Future research should be focused on finding new formulations, technological approaches and process parameters that will allow to attenuate the negative impact caused by those.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

AUTHOR CONTRIBUTIONS

All authors contributed equally to this work.

FIGURE CAPTION

Figure 1. Example of by-products reuse to produce gluten-free products with a circular economy approach.

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Table 1. Main functional molecules in plant-based waste and by-products used in gluten-free foods.

Waste and By-products	Functional molecules	References
Fruits		
Green banana and peel	Dietary fibre, carotenoids, vitamins, catechin, epicatechin, gallocatechin	Zandonadi et al.[28] Khoozani et al.[27]
Passion fruit peel	Pectin, cellulose and hemicellulose	Bussolo et al. [30] Coelho et al. [31]
Orange pomace	Dietary fibre, minerals (calcium, magnesium and potassium)	O'Shea et al. [32] Kırbaş et al. [33]
Apple pomace	Dietary fibre, minerals (potassium and iron), proanthocyanidins, hydroxycinnamates, flavonols, dihydrochalcones and anthocyanins	O'Shea et al. [32] Kırbaş et al. [33] Waldbauer et al. [34]
Coconut oil waste	Dietary fibre, fat and minerals (calcium, sodium and magnesium)	Widiastuti et al.[36]
Pomegranate seeds	Polyunsatured fatty acids (punicic, linoleic, oleic) and ellagic, chinic and malic acids	Bustamante et al. [40] Rouhi et al. [43]
Olive leaves and mill wastewater	Oleuropein, hydroxytirosol, hydroxytirosol glucoside, tyrosol, caffeic, coumaric and ferulic acids	Difonzo et al. [45] Paulo et al. [46]
Grape peel	Dietary fibre, sugar, minerals (potassium, magnesium and calcium) phenolic compounds	Iuga et al. [49]
Strawberry, blueberry, chokeberry, blackcurrant and raspberry seeds	Dietary fibre, protein and polyphenols (mainly anthocyanins)	Sójka et al. [52] Struck et al. [51]
Coffee husk and silverskin	Dietary fibre, protein, minerals, tannins, cholorogenic acid and caffeine	Iriondo-DeHond et al. [54] Janissen et al. [53]
Vegetables		
Carrot pomace	Dietary fibre, phenolic compounds, carotenoids, uronic acids and sugars	Surbhi et al. [55]
Broccoli leaves	Protein, minerals, and glucosinolates.	Domínguez-Perles et al. [59] Drabińska et al. [61]
Potato peel	Caffeic, gallic, protocatechuic and chlorogenic acids	Singh et al. [63]
Tiger nut milk by-product	Dietary fibre	Sánchez-Zapata et al. [68]
Cereals, oilseed and legumes		
Rice bran	Dietary fibre, protein, minerals (phosphorus), vitamins, phytosterols, oryzanols and tocotrienols	Sohail et al. [70] Sharif et al. [69]
Chia seed cake	Dietary fibre, protein, polyphenols	Hessle et al. [72] Aranibar et al. [73]
Hemp seed cake	Dietary fibre, protein and essential amino acids (edestin and albumins)	Pojić et al. [75]
Okara	Dietary fibre and protein	Lu at al. [77]
Carob's germ and seed	Dietary fibre, protein, hydrozybenzoic acids, gallotannins, flavones and flavonols	Rico et al. [78]
Chickpea hull	Dietary fibre, carotenoids, anthocyanins	Costantini et al. [79]

Table 2. Effect of by-products addition on physico-chemical and sensory parameters of gluten-free products.

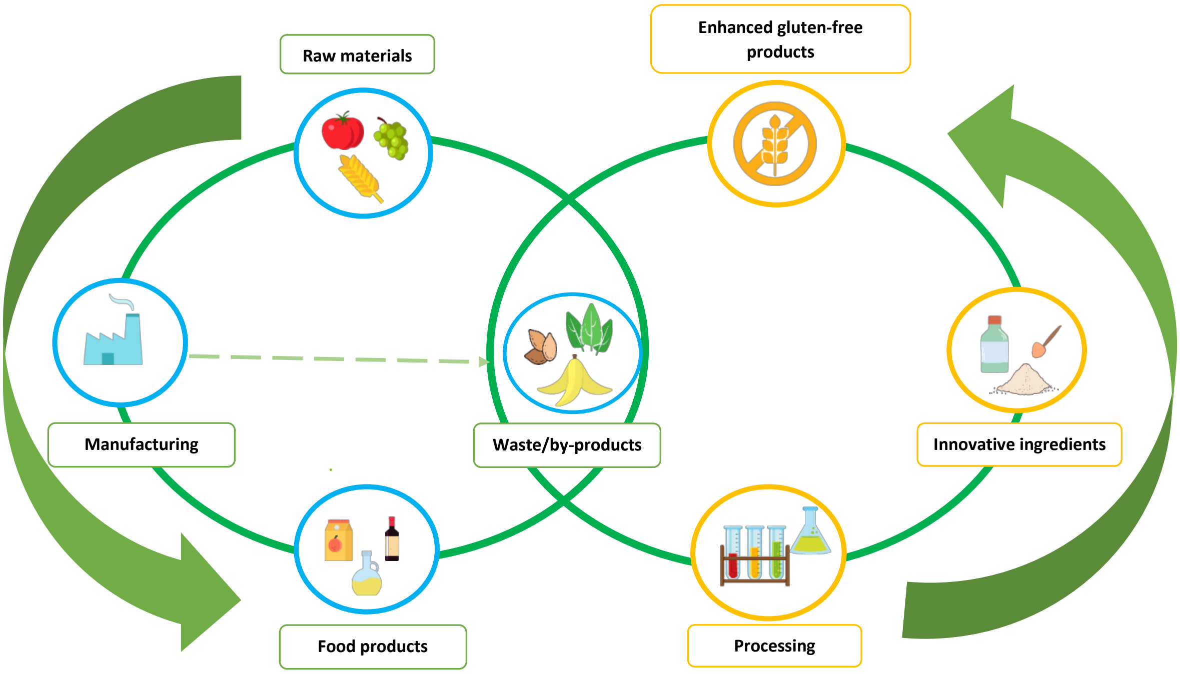
By-Products	Amount	GF product	Effects	References
Apple pomace	5-20%	Bread	↑crumb hardness, ↓cohesivness, resilience,↑a^* and b^* value.	Rocha Parra et al. [82]
Orange pomace	0-8%		↓crumb hardness with low levels of orange pomace, ↑firmness, crumb density, ↓specific volume high levels of orange pomace	O'Shea et al. [84]
Rice bran	5-20%		↓firmness, ↑colour intensity, ↑bread specific volume	Genevois et al. [86]
Tiger nut milk by-products	5%		↓bake loss, ↑hardness, ↓sensory acceptability	Aguilar et al. [87]
Carrot pomace	10-30%	Cake	↓density, hardness, cohesiveness, ↑symmetry index, sensory score	Majzoobi et al. [89]
Black carrot pomace	3-9%		↓L^*, b^*, water activity, ↓specific volume, firmness, cohesiveness and gumminess, ↑ overall acceptability	Singh et al. [90]
Apple pomace	18.15/18.28%		↓hardness, springiness, cohesiveness and gumminess, ↑taste and colour	Azari et al. [91]
Green banana peel	5-20%		↓volume, height, baking loss	Türker et al. [93]
Broken bean	45-75%		↓specific volume, ↑firmness, ↓L^*,↑a^*, ↓b^*, ↓appearance	Gomes et al. [94]
Grape and pomegranate seeds	5%		↑hardness, ↓symmetry index, ↓L^*, ↓b^*, ↓mouthfeel	Levent et al. [95]
Orange peels	4.5%	Muffin	↑hardness, chewiness, ↓colour homogeneity, ↑sensory acceptability	Talens et al. [92]
Potato pulp	90-75%	Pasta	↓cooking time, solid loss, ↑yield, ↑$b^*$and a^* value	Bastos et al. [99]
Passion fruit peel	10/20%		↑cooking time, soluble solids loss and water absorption, ↓L^*, a^*, ↓overall appearance, taste and colour	Ribeiro et al. [100]
Durian seed	25/50%		↑cooking yield, ↑a^*, ↓$L^*$and b^*, ↓adhesiveness and overall acceptability, ↑hardness	Mirhosseini et al. [101]
Green banana and green banana peel	12.5%		↓cohesiveness, chewiness, elasticity	Patiño-Rodríguez et al. [102]
Broken kernels	10-25%		↓cooking loss, stickiness, adhesiveness, ↑water absorption and firmness with 20% of rice gel	Fradinho et al. [103]
Passion fruit peel	3%	Breakfast cereals	↑a^*, ↑expansion index and specific volume	Alonso et al. [104]
Olive leaf and mill wastewater	0.05-0.1%	Breadstick	↓a^*, ↓hardness	Conte et al. [105]

↓= decrease; ↑= increase

Table 3. Effect of by-products addition on nutritional parameters of gluten-free products.

By-Products	Amount	GF Product	Effects	References
Okara	15-50%	Cookies	↑protein, fibre and ash, ↓ caloric value	Ostermann-porcel et al. [106]
Orange pomace	15-35%		↑fibre, protein, ash and minerals (potassium, phosphorus, calcium, iron, copper, manganese and sodium)	Cayres et al. [107]
Defatted strawberry & blackcurrant seeds	5-15%	Bread	↑fibre, protein, polyphenols and antioxidant activity	Korus et al. [110]
Pressing of chia seed oil	5%		↑ash, polyphenols and antioxidant activity, ↓energy value	Zdybel et al. [123]
Coffee silverskin and husk	2.5%		↓carbohydrates, lipids, ↑protein, fibre, polyphenols, chlorogenic acid and antioxidant activity	Guglielmetti et al. [125]
Coffe cascara	3-4.5%		↑fibre and protein	Rios et al. [126]
Pomegranate seed	2-10%		↑polyphenols and antioxidant activity	Bourekoua et al. [131]
Carrot pomace	5-15%	Muffins	↑polyphenol, caroteinoid and antioxidant activity	Olawuyi et al. [111]
Green banana	47%	Pasta	↓lipids and caloric value, ↑ash	Zandonadi et al. [28]
Green banana	15-50%	Cakes	↓protein, lipids, ↑fibre and potassium	Segundo et al. [112]
Broccoli leaf	2.5-7.5%		↑glucosinolate, total phenols and antioxidant activity	Drabinska et al. [61]
Cactus peel	5-10%	Snacks	↓fat, moisture	Miranda et al., [113]
Green banana	10-50% 5-25%		↑fibre, essential minerals (potassium, calcium, magnesium and phosphorus), polyphenol and antioxidant activity, ↓oil content	Wang et al. [118]
Hemp seed oil press-cake	10-40%		↑protein, fibre, omega-3 fatty acids and antioxidant activity	Radočaj et al. [119]
Virgin coconut oil waste	10-50%	Flakes	↑protein, fibre, fat, calcium and iron, ↓sodium	Widistatu et al. [36]

↓= decrease; ↑= increase



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964 **Figure 1**