

1 Review

2 Carob bean (*Ceratonia siliqua L.*): a new perspective  
3 for functional food

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5 Pintado.

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13 free, fibres, gastrointestinal benefit.

14 **Statistical summary:** 8938 words, 4 tables and 2 figures.

15 **Abbreviations:** Locust bean gum (LBG), carob fruit extract (CFE), carob pods aqueous  
16 extract (CPAE), inositol-enriched beverage (IEB), reactive oxygen species (ROS),  
17 reactive nitrogen species (RNS), total cholesterol (TC), low-density lipoprotein  
18 cholesterol (LDL), very low-density lipoprotein cholesterol (VLDL), triglycerides (TG),  
19 cardiovascular disease (CVD), gastrointestinal disorders (GID), gastrointestinal transit  
20 (GIT), 1,2-dicarbonyl compounds (DCs), hydroxymethylfurfural (HMF).

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38 **ABSTRACT**

39 *Background*

40 Carob (*Ceratonia siliqua* L.) is an evergreen tree that belongs to the Leguminosae family  
41 and is typical of the Mediterranean basin. It is well known for its valuable locust bean  
42 gum obtained from carob seeds. However, the food industry can obtain different carob  
43 products from carob fruit after processing. Carob products are good sources of dietary  
44 fibre, sugars, and a range of bioactive compounds such as polyphenols and D-pinitol.

45 *Scope and Approach*

46 Bioactive compounds present in carob fruit and its derived products help control many  
47 health problems such as diabetes, heart diseases, and gastrointestinal disorders due to their  
48 anti-hyperglycaemic, antioxidant, and anti-inflammatory activities. So, carob products  
49 have a great potential to be used as a functional food ingredient.

50 *Key Findings and Conclusions*

51 This article focuses on carob characteristics and processing, chemical composition, health  
52 benefits, and applications in food formulations to explore the potential of carob in  
53 developing a wide variety of health-beneficial food products.

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## 61        **1. Introduction**

62    Less known fruit species, including carob, are gained more popularity recently. They  
63    include a high content of non-nutritive, nutritional, and bioactive compounds such as  
64    flavonoids, phenolics, anthocyanins, phenolic acids, and as well as nutritional compounds  
65    such as sugars, essential oils, carotenoids, vitamins, and minerals. Less known fruits have  
66    distinct flavour and taste, excellent medicinal value, and health care functions as well  
67    (Engin & Mert, 2020; Eydurán et al., 2015; Gundogdu et al., 2014; Kaskoniene et al.,  
68    2020).

69    Carob (*Ceratonia siliqua* L.) is an evergreen tree that belongs to the Leguminosae or  
70    *Fabaceae* family (subfamily *Caesalpinioideae*) native to the Mediterranean region. It has  
71    been widely exploited since antiquity due to its edible fruits (commonly referred to as  
72    pods or merely carob). It is still currently used in agro-food industries and soil restoration  
73    purposes (Tous & Antoni, 2013). In the last years, the average production of carob pod  
74    in the world was reduced from 165,990 tonnes in 2013 (Yatmaz & Turhan, 2018) to  
75    136,612.75 tonnes in 2018 (FAOSTAT, 2020). The fall of carob cultivated hectares in  
76    important producers such as Spain explains these figures. Carob trees have been  
77    traditionally used to obtain locust bean gum (LBG; thickener E410) from the pod seeds  
78    that accounts for only 10% of the pod's weight. The emergence of cheaper substitutes for  
79    LBG, such as guar or xanthan gum, made carob tree cultivation no longer profitable in  
80    the EU (López-Sánchez, et al. 2018). However, many countries still cultivate carob trees,  
81    which is justified by pharmacological and food industry interests. The main worldwide  
82    producers of carob pod from 2015 to 2018 were Portugal (28.83%), Italy (23.11%),  
83    Morocco (16.11%), and Turkey (10.39%) (FAOSTAT, 2020)(Table 1).

84    The four main genetic groups of *C. siliqua* identified are SM, South Morocco; SS, South  
85    Spain; CM, central Mediterranean including genotypes from Portugal, Algeria, France,

86 Sardinia, Sicily and Balearics; EM, eastern Mediterranean (*i.e.* Greece, Cyprus and  
87 Lebanon) (Viruel et al., 2019). The use of carob dates to the ancient Egyptians who fed  
88 livestock with carob pods and are also reputed to have used the gum as an adhesive in  
89 mummy binding. The Arabs used the carob seed as a unit of weight. They called the seed  
90 *qirat* or *karat*, and the standard weight of the carob seed became the unit of weight for  
91 gold and precious stones. At present, carob pods are commonly used in cakes, cookies,  
92 drinks, and various snacks in Egypt. Jams and liquors are made from carob in Turkey,  
93 Malta, Portugal, Spain, and Sicily. In Libya, a syrup named *rub* is extracted from carob  
94 and used to make *asida*, a traditional dessert.

95 The carob tree is a xerophytic (drought-resistant) species, well adapted to the  
96 Mediterranean region's ecological conditions with deep root systems, which are intolerant  
97 to waterlogging (Gucel & Sakcali, 2012). Given the rising climatic crisis, this plant  
98 requires little maintenance and can be a good solution in the future, both ecologically and  
99 economically. Santos et al. (2019) reported high accuracy the thermal growing conditions  
100 of carob tree cultivated in the Algarve region, a limited area with a well-defined climate  
101 in Portugal. This study confirms that carob tree is better located in warm areas with low  
102 chill requirements, where temperatures ranged between 16 to 36 °C for more than 50%  
103 of the hours of a year. This feature is one reason why the possible role of carob tree in the  
104 fire protection of agro-forest ecosystems of Mediterranean countries is considered.  
105 Besides, it is an essential component of Mediterranean vegetation, that in the absence of  
106 pruning practice, it is converted into bush-like growing formations. Another reason is  
107 their easy planting propagation (Srećec et al., 2017). This knowledge is of great  
108 importance for different socio-economic sectors, such as agriculture and forestry, e.g., by  
109 assisting bioclimatic zoning and suitability assessments.

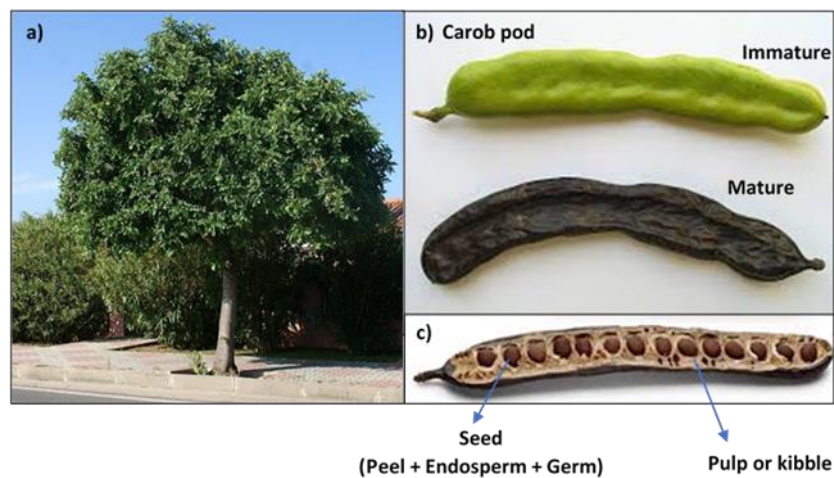
110 Carob has a great potential to be used in the food industry, not only for its health benefits  
111 (Goulas et al., 2016) and its characteristic strong aroma, which remains even after  
112 processing. This unique property may be explained by the presence of acids, esters and  
113 aldehydes/ketones emitted from carob fruit and powder, which are biogenic volatile  
114 organic compounds that contribute to plant growth, breeding and defence (Krokou et al.,  
115 2019). On the other side, the food industry's diversification and innovative development,  
116 brand new technological modernisation, and product-line expansion strategies are  
117 needed. Lobanov et al. (2018) studied the economic impact of incorporating vegetal  
118 ingredients in the innovative production of flour-based functional foods. In particular, the  
119 production of a lecithin bun with 4% of carob resulted in an increased content of proteins,  
120 fat and fibres and a decreased carbohydrate content, with better production efficiency than  
121 the production of lecithin bun without carob. The price of the carob bun is only 6% higher  
122 than the one without carob addition. This analysis allows recognising carob bean as an  
123 excellent raw material to produce gluten-free bread and flours enriched with vitamins,  
124 minerals, and proteins.

125 This work aims to review and highlight the beneficial aspects of carob and its potential  
126 for use as a functional ingredient in the food industry, opening new perspectives for this  
127 sustainable crop.

## 128 **2. Carob characteristics and processing**

129 Functional food is defined as a food or beverage product containing nutraceutical or food-  
130 grade bioactive agents with health benefits over and above normal nutritional function  
131 (McClements & Xiao, 2014). Nowadays, there is a growing interest in supplements  
132 derived from natural, traditional and non-traditional foods as possible sources of  
133 biologically active substances with proven health properties for inclusion in the human  
134 diet (Baumel et al., 2018). About that, carob (*Ceratonia siliqua* L.) holds potentially

135 significant importance for the food industry due to its phytochemical constituents with  
 136 functional properties (Goulas et al., 2016), flavouring properties (Foundation & Aue,  
 137 1997), and nutrition benefits (Papaefstathiou & Agapiou, 2018). The carob tree grows up  
 138 to 8–17 m high, with a broad semispherical crown and a thick trunk with brown, rough  
 139 bark and sturdy branches (Figure 1a; Tous & Antoni, 2013). The tree produces fruit in an  
 140 edible bean/pod, which is also known as locust bean. The beans (pods) hang in the form  
 141 of clusters and remain green until fully matured (Figure 1b). The mature fruit is a long  
 142 scimitar-shaped pod (10 to 25 cm) and contains several hard seeds embedded in a pulp  
 143 (Figure 1c). The pods vary significantly in morphological characteristics such as  
 144 dimension, size, weight, shape, density, colour, and seed-to-pulp ratio due to differences  
 145 in cultivar varieties and climatic conditions (Benchikh et al., 2014; Nasar-abbas et al.,  
 146 2016; Tous & Antoni, 2013). The seed:pulp ratio can vary between 10-20%:80-90%,  
 147 increasing from cultivated type to wild type, because the pulp part of the wild type is  
 148 smaller than the cultivated one (Yatmaz & Turhan, 2018). Carob seeds are characterised  
 149 by a brown colour, significant hardness, a length around 10 mm, and a weight of about  
 150 0.2 g per seed. Seeds are composed by peel (30–33% w/w), endosperm (42–46% w/w),  
 151 and germ (23–25% w/w) (Figure 1c; Tous & Antoni, 2013).

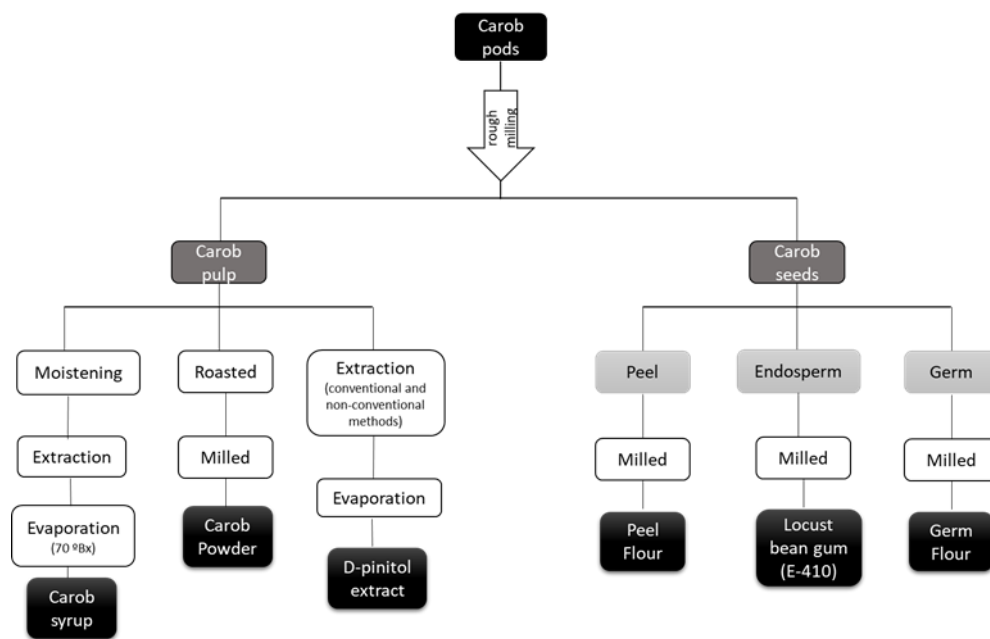


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153 Figure 1. a) Carob tree (*Ceratonia siliqua* L.), b) Immature and mature carob pod, c)  
 154 Carob pod constituents (Tous & Antoni, 2013).

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156 Carob pods are rich in sugars (45-52%) and crude fibre (up to 40%) but poor in protein  
157 (2-7%) (Nasar-abbas et al., 2016). Most of the protein is found in the seed germ (55-65%)  
158 (Dakia et al., 2007), and the seed endosperm contains a high percentage of galactomannan  
159 (80-85%) (Barak & Mudgil, 2014). Due to the differences in the composition and  
160 functional properties, carob pulp and seeds are used separately to produce many products.  
161 The main industrial uses of the carob fruit are shown in Figure 2.



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163 Figure 2. Main industrial uses of carob fruit (Durazzo et al., 2014; Ersan et al., 2020;  
164 Tounsi et al., 2017; Yousif & Alghzawi, 2000).

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166 The manufacture of primary carob products such as flour, powder, and syrup requires pre-  
167 separation of seeds from the pod (Loullis & Pinakoulaki, 2018). The carob pods are dried  
168 after harvesting to reduce moisture to around 8%, and subsequently, they are kibbled to  
169 separate the seeds from the pulp portion. The pulp is processed differently, depending on  
170 whether it will be used for animal feed or food products. For animal feed, the pulp is  
171 grounded into different sizes based on the kind of livestock to be fed. In the food industry,  
172 the pulp is processed to obtain carob syrup and carob powder (Tous & Antoni, 2013).  
173 Carob syrup is obtained by leaving the grounded carob pulp in water. The mixture is then

174 drained and boiled until desirable consistency (Özcan et al., 2009). Aziz & Hicham (2014)  
175 optimised the carob syrup production from different populations of Moroccan carobs. The  
176 authors used an experimental design to investigate the effect of three parameters  
177 (extraction temperature, extraction time and ratio of water to pulp) on syrups yields. As  
178 results, the optimum conditions were extracting temperature of 43.45 °C, extracting time  
179 of 2.40 h and extraction ratio solvent (water) to the pulp of 2.27 (v/w). The syrup yield  
180 from the different regions varies between 28.76 and 37.22 g/100 g (% w/w). The  
181 production and purification of sugar syrup can also be performed by a 10 column system  
182 connected in series, reaching a syrup yield of at least 90% (Petit & Pinula, 1995).

183 The pulp is first dehulled for carob powder production, grounded to different sizes,  
184 roasted and finally milled into a fine powder, called carob flour or powder (Yousif &  
185 Alghzawi, 2000). Based on the time and temperature used for roasting, lightly, medium,  
186 and highly roasted carob powder can be obtained. Generally, in conventional roasting of  
187 carob pulp, a temperature range of 120–180 °C (mostly around 150 °C) is used for 10–60  
188 min.

189 Additionally, D-pinitol (3-o-methyl-d-chiro-inositol) extract can be obtained from carob  
190 pulp. This natural compound, related to the important family of inositols, is present in  
191 carob in high levels and increases this *Leguminosae*'s value due to its pharmacological  
192 importance related to anti-cancer, anti-diabetic, antioxidant, and anti-ageing properties  
193 (López-Sánchez et al., 2018). Besides the classic extraction associated with the syrup  
194 carob production, D-pinitol extraction from carob pulp was studied using microwave  
195 (Ersan et al., 2020), supercritical CO<sub>2</sub> (Cháfer & Berna, 2014) and ultrasound (Tetik &  
196 Yüksel, 2014) techniques.

197 Finally, the carob seed consists of three parts: the peel (husk), the endosperm and the  
198 germ. At first, the seeds are dehusked by treatment with dilute sulphuric acid or with a



199 thermo-mechanical treatment known as acid peeling and thermal peeling, respectively  
200 (Prajapati et al., 2013). The isolated endosperm is then milled and sieved to obtain fine  
201 particle size powder of native locust bean gum (LBG), a polymer used as a natural food  
202 additive, and several applications in the pharmaceutical industry (Barak & Mudgil, 2014).  
203 The LBG produced from the acid peeling treatment is whitish and has a high viscosity.  
204 Simultaneously, the one from thermo-mechanical peeling is somewhat darker due to  
205 heating or roasting operations. The produced LBG can be further clarified by dissolving  
206 in water under heating conditions. The solution is filtered to remove insoluble substances  
207 and then dried and milled to obtain clarified or purified LBG (Tous & Antoni, 2013).

### 208 **2.1. Carob pulp chemical composition**

209 The chemical composition of carob pulp varies with genetic, environmental, climatic  
210 factors, and harvesting season. The plant type (male, female, or hermaphrodite) and  
211 cultivar significantly influence carob pulp's chemical composition and biological  
212 activities. Simsek et al. (2017) collected the carob fruits (wild and cultivated) from  
213 Mersin in Turkey at different harvest periods (May to August) and analysed the amino  
214 acid and sugar contents. Protein content ranged from 6.09% (cultivated) to 9.08% (wild),  
215 depending on the harvest periods. Total amino acid contents ranged between 3.87 and  
216 8.21%. Cultivated carob harvested in May showed the highest amount of aspartic acid  
217 (1.28%). The authors found the highest fructose and glucose contents (40.26 and 17.69%)  
218 in wild carob fruit harvested in the June period. They also found the sucrose content of  
219 wild and cultivated carob fruit at the latter harvested to be highest (30.58 to 30.08% and  
220 31.42 to 30.77%, respectively). In another study, carob fruit contained 6.90–7.44%  
221 fructose, 2.0–2.26% glucose, and 32.6–45.4% sucrose (Khelifa et al., 2013). Avallone et  
222 al. (1997) reported moisture (6–10%), ash (1–6%), protein (1–5%), fat (0.4–0.8%),  
223 sucrose (27–40%), D-glucose (3–5%), D-fructose (3–8%) and starch (0.1–1.3%) contents

224 (in dry weight) of carob pod samples from eastern parts of Sicily in Italy. In the same  
225 work, the total polyphenol (15.8-24.4 mg/g), condensed tannins (proanthocyanidins:  
226 2.09-3.89 mg/g), and hydrolysable ones (ellagitannins: 0.34-0.68 mg/g and gallotannins:  
227 0.2-0.6 mg/g) contents were also reported. There is great variability of phenolic and tannic  
228 fractions in carob pod samples (Avallone et al., 1997). Özcan et al. (2009), Oziyci et al.  
229 (2014) and Ayaz et al. (2007) reported minerals contents of the carob fruit, being  
230 consensual that major proportions are of potassium (970 to 1089 mg/100 g), calcium (266  
231 to 319 mg/100 g), phosphorous (76 to 79 mg/100 g), and magnesium (55 to 56 mg/100  
232 g). Carob pulp also contains appreciable amounts of dietary fibre, principally insoluble  
233 fibre (68.4%) (Owen et al., 2003). It is almost free from stimulants such as caffeine and  
234 theobromine (Loullis & Pinakoulaki, 2018).

235 All these phytochemicals represent an essential source of nutrients and compounds that  
236 are beneficial to human health. Consequently, traditional products produced from carob  
237 pulp, such as syrup and powder, can be considered functional food products.

### 238 **2.1.1. Syrup**

239 Özcan et al. (2009) studied the composition and mineral contents of carob syrup.  
240 According to these authors, the product does not contain oil and had higher moisture  
241 values (34.65%), total sugar (63.88%), and higher water and alcohol solubility when  
242 compared to the carob fruit. The syrup has the highest values regarding mineral  
243 composition (potassium, phosphorus, and calcium). Besides, carob syrup has higher  
244 levels of silver and titanium than carob fruit, and it does not contain zinc. These  
245 physicochemical characteristics allow considering carob syrup as a functional ingredient.  
246 Fidan et al. (2019) evaluated the physicochemical characteristics of sponge cakes  
247 enriched with carob syrup. They proved that the replacement of sugar with carob syrup  
248 increased the protein content level (from 8.44 to 12.57g/100 g in dry weight) but did not

249 observe significant changes in the carbohydrates and dietary fibres contents. They  
250 previously evaluated the content of reducing sugars and total carbohydrates in carob pulp  
251 before and after extraction of syrups (Fidan et al., 2016). During carob processing,  
252 increases in reducing sugars were observed and could be from carbohydrate hydrolysis  
253 during storage or heat treatment during syrup production. The carbohydrate content in the  
254 syrup prepared from Turkish carob pods was highest, with the sucrose content especially  
255 reaching up to 45 g/100 g dry weight compared with the sucrose content in carob pulp  
256 (34.2 g/100 g dry weight).

257 Concerning the natural phenolics contents of carob, Dhaouadi et al. (2014) suggested that  
258 carob syrups exhibit antioxidant, antibacterial and antifungal activities affected by sugar  
259 supplementation syrup processing. Avoiding sugar addition during syrup preparation  
260 preserves carob syrup bioactivities and prevents negative nutritional attributes since  
261 sucrose is associated with physiological disorders (mainly diabetes and obesity).

262 Additionally, it is known that carob is the only raw material from which D-pinitol can be  
263 isolated in quantities enough for possible commercial exploitation (López-Sánchez et al.,  
264 2018). Tetik et al. (2011) determined the D-pinitol content of 10 different carob syrup  
265 samples purchased in Turkey. D-pinitol means the content was found to be  $84.63 \pm 10.73$   
266 g/kg dry weight. Christou et al. (2019) developed a gas chromatography-mass  
267 spectrometry (GC-MS) method to quantify D-pinitol in carob syrup samples with origin  
268 in Cyprus. D-pinitol was determined in 13 carob syrup samples, and the mean  
269 concentration was  $68.58 \pm 4.80$  g/kg dry weight. Compared to other plants or legumes,  
270 carob appears to be the richest source of D-pinitol, highlighting carob's role as a  
271 functional organic food (López-Sánchez et al., 2018).

### 272 **2.1.2. Powder**

273 The chemical composition of carob powder is given in Table 2, however it varies with  
274 genetic, environmental and climatic factors (Faik A. Ayaz et al., 2009; Carbas et al., 2019;  
275 Özcan et al., 2009; Petkova et al., 2017). To obtain powder or flour from carob fruit, a  
276 roasted, milled, and sieved processing must be carried out. Conventional roasting (with  
277 hot air) has been used since ancient times. However, and recently, some works  
278 approached the use of the microwave for roasting, concluding that this is a less time and  
279 energy-consuming method, allowing to obtain a final product with a higher nutritional  
280 profile when compared to the one obtained with conventional heating (Gunel et al., 2018,  
281 2019). The roasting step is crucial to attaining the product's stability throughout storage,  
282 affecting its chemical composition (Boublenza et al., 2017; Červenka, et al., 2019; Vitali  
283 Čepo et al., 2014). In this last aspect, sugar caramelisation and Maillard reaction favour  
284 the production of furans, esters and pyrroles that recall cocoa's aroma (Foundation & Aue,  
285 1997). Moreover, the release of phenolics occurs through the ruptured of high-molecular  
286 complexes from the carob matrix or the partial degradation of phenolics, which results in  
287 the production of different types of antioxidant molecules (Boublenza et al., 2017). These  
288 phenolic compounds and Maillard reaction products contribute to carob products'  
289 beneficial health properties such as antioxidant capacity.

290 The chemical characteristics of carob powders are significantly influenced by the plant's  
291 variety or geographical origin and the carob pod's ripening stage (Farag et al., 2019; Srour  
292 et al., 2016). Rodríguez-Solana et al. (2019) demonstrated that the carob roasting degree  
293 and variety have an apparent positive effect on total phenolics content and antioxidant  
294 activity. Özcan et al. (2009) roasted, milled and sieved carob fruit obtained from local  
295 carob-processing plants in Antalya, Turkey, and compared the chemical composition and  
296 mineral content of carob flour the ones of the carob fruit. Protein, oil, crude fibre, ash and  
297 energy values of carob fruit and carob flour were not statistically different. Still, the water

298 solubility of carob flour was lower than the solubility of the fruit. Carob flour contains  
299 potassium, phosphorus, calcium, and sodium in higher amounts regarding the mineral  
300 composition.

301 Furthermore, the contents in silver, aluminium, barium, iron, and zinc determined in the  
302 carob flour were the highest ones. Youssef et al. (2013) studied the chemical composition  
303 of carob powder obtained from Aswan, Egypt. They concluded that the powder is also  
304 rich in vitamins E, D, C, Niacin, B6 and folic acid. In Faro, Portugal, Carbas et al. (2019)  
305 reported that commercially available carob flours had high dietary fibre levels, total  
306 phenols, D-pinitol, monosaccharides, and antioxidant activity. Another study from  
307 Turkey (Fidan et al., 2018) demonstrated that carob flour had high amounts of protein  
308 (22.56%) and dietary fibre (28.17%), being a good source of antioxidants, particularly  
309 polyphenolic compounds and minerals (Mg, Fe, and Zn). These phytochemicals have  
310 important healthy and nutritional properties that corroborate the use of carob flours as  
311 food ingredients. Applying carob flour as a cocoa substitute in the confectionery (Loullis  
312 & Pinakoulaki, 2018) or beverages and mixtures with other cereals-based products are  
313 examples.

## 314 **2.2. Carob seeds chemical composition**

315 As previously mentioned, the main use of carob seeds is LBG production, a popular  
316 natural polymer consisting of galactose (G) and mannose (M) in the ratio 1:4 and hence  
317 known as galactomannan (Kapoor & Divisions, 2005). LBG is obtained by grinding the  
318 endosperm layer after dehulled without damaging the endosperm and the germ (Prajapati  
319 et al., 2013). The major portion of LBG endosperm is galactomannan, which comprises  
320 approximately 80% of the endosperm weight, corresponding the remaining part to  
321 proteins and impurities. The commercial LBG powder contains approximately 10–12%  
322 moisture, 5% protein, 1.0% ash, 1.0% crude fibre, 0.5% fat, and 80–85% galactomannan

323 of which 50-65% is mannose, 14-18% is galactose and the rest are traces of glucose,  
324 rhamnose, arabinose and xylose (Barak & Mudgil, 2014). LBGs can differ in their G:M  
325 ratio, chemical composition, and physicochemical properties such as solubility, molecular  
326 size and dynamic viscosity. Those depend on the origin, variety and age of the carob tree,  
327 growth conditions (*e.g.*, climate and soil) and polysaccharide extraction method used for  
328 crude gum purification (Bouzouita et al., 2007; Haddarah et al., 2014; Smith et al., 2008).  
329 LBG is widely used as a thickener and stabiliser with numerous applications in the food,  
330 pharmaceutical, cosmetic, and biotechnology industries (EFSA Panel on Food Additives  
331 Nutrient Sources added to Food et al., 2017). It is a globally approved food additive  
332 (E410) with proven enhanced properties used in different food matrixes. This gum is used  
333 as an edible film/coating to increase the shelf life of fresh fruits, vegetables, and meat  
334 products. Additionally, it is a relevant constituent of many products such as beverages,  
335 yoghurt, noodles, puddings, cheeses, water-based jellies, candies, fish products, ketchup,  
336 mayonnaise, bakery products, and frozen foods to improve texture properties (Barak &  
337 Mudgil, 2014).

338 With the large quantities of LBG being produced annually, an appreciable amount of  
339 carob germ flour is coproduced as a by-product of the process (Smith et al., 2008). Dakia  
340 et al. (2007) studied the dehulling procedure's influence on the germ meal composition,  
341 observing small reductions in protein and lipid contents from acid extraction. The carob  
342 germ (consisting of fine fragments of hull and endosperm), which could be obtained  
343 industrially, is 8.3% moisture, 6.5% ash, 6.6% lipids (neutral and polar) containing  
344 approximately 21% polar lipids, 54.7% crude proteins, and has an energy value of 17.5  
345 kJ/g (Dakia et al., 2007). The composition and molecular weight distribution of carob  
346 germ protein were analysed by Smith et al. (2008). They used the modified Osborne  
347 extraction procedure (Osborne, 1907) to divide proteins into the following solubility

348 classes: water and salt soluble proteins (albumins and globulins), aqueous alcohol-soluble  
349 (nonreduced) proteins (prolamins), insoluble aqueous alcohol-soluble (reduced) proteins  
350 (cross-linked prolamins), and alkali-soluble proteins (glutelins). They determined that the  
351 carob germ flour proteins contained albumin and globulin (ca. 32%) and glutelin (ca.  
352 68%) with no prolamins detected, which determines the gluten-free concept (Cebolla et  
353 al., 2018). Prolamins have been widely studied due to their role in the rheological  
354 characteristics of bakery and pasta products made from wheat flour (Mamone et al., 2011;  
355 Martínez-Esteso et al., 2016). Rice & Ramstad (1950) compared the amino acid  
356 composition of gluten to carob proteins. They found significant differences between the  
357 two proteins, with carob germ proteins having less cysteine, glutamic acid, phenylalanine,  
358 and more of the charged amino acids, arginine, aspartic acid lysine. Feillet & Roulland  
359 (1998) isolated proteins from carob germ flour as described by Rice & Ramstad (1950)  
360 and designated these proteins as “caroubins”. Feillet & Roulland (1998) concluded that  
361 the ability of caroubin to form a viscoelastic material might result from protein  
362 interactions. Components such as lipids and carbohydrates may also contribute to the  
363 viscoelastic properties of the material. Bengoechea et al. (2008) also studied the amino  
364 acid composition and the nature of the subunits that compose carob germ proteins. These  
365 authors obtained 96.5% of protein content by alkaline extraction, followed by isoelectric  
366 precipitation of proteins. A high number of amino acids like glutamic acid, aspartic acid  
367 and arginine was detected. Carob proteins were composed of aggregates formed by 131  
368 and 70 kDa subunits linked by non-covalent bonds and other peptides strongly bounded  
369 by disulfide interactions. Both aggregates and subunits were formed mainly by 100 and  
370 48 kDa monomers linked by disulfide bonds. A considerable content of high molecular  
371 mass peptides strongly bounded were also found. Proteins became partially denatured and  
372 thermally stabilised at acidic conditions (pH=2). Furthermore, the relatively high content

373 of phospholipids (11.8%, v/v) might increase the antioxidant potential and improve the  
374 baking performances of flour fortified with carob seed germ (Siano et al., 2018).

375 The carob seed peels extracts obtained by sequential extraction with solvents with  
376 different polarities were studied by Lakkab et al. (2019). These authors concluded that  
377 carob seed peel has high phenolics contents and strong antioxidant activity. They assessed  
378 its effects on mood disorders and anxiety, opening new mechanistically approaches to  
379 antidepressant and anxiolytic drug development.

380 More recently, Fidan et al. (2020) evaluated the chemical composition, antioxidant  
381 potential and functional properties of carob seeds. They concluded that carob seeds are a  
382 valuable source of phenolic compounds and antioxidants. Carob seeds also presented high  
383 proportions of proteins, lipids, and galactomannan with functional properties (good  
384 swelling properties and oil-holding capacity) that could improve the nutritional value of  
385 foods in which they are incorporated. Furthermore, the seeds have high concentrations of  
386 macro and micro minerals such as Ca, Fe, Mg, and Zn (Oziyici et al., 2014).

### 387 **3. Health benefits**

388 Numerous studies have revealed physiological responses to carob fruit and its products  
389 that may be considered health beneficial by healing and preventing chronic diseases. The  
390 carob chemical components and their biological properties associated with health benefits  
391 are presented in Table 3.

#### 392 **3.1 Antioxidant properties**

393 Reactive oxygen species (ROS) and reactive nitrogen species (RNS) are a wide variety  
394 of free radicals and ions. They are produced either from normal cell metabolisms *in situ*  
395 (cellular redox process) or from external sources (pollution, cigarette smoke, radiation,  
396 medication, alcohol, cooking). These species play a dual role as both toxic and beneficial  
397 compounds. The delicate balance between their two antagonistic effects is an essential



398 aspect of life. At low or moderate levels, ROS and RNS exert beneficial effects on cellular  
399 responses and immune function. At high concentrations, they generate oxidative stress, a  
400 deleterious process that can damage all cell structures. Oxidative stress plays a significant  
401 part in developing chronic and degenerative ailments such as cancer, arthritis, ageing,  
402 autoimmune disorders, cardiovascular and neurodegenerative diseases. The human body  
403 has several mechanisms to counteract oxidative stress: (i) by producing antioxidants,  
404 which are either naturally produced *in situ* that can be classified as enzymatic antioxidants  
405 (superoxide dismutase, catalase, glutathione peroxidase and glutathione reductase), or  
406 non-enzymatic antioxidants (lipoid acid, glutathione, L-arginine, coenzyme Q10,  
407 lactoferrin, etc.); (ii) or externally supplied through foods and/or supplements such as  
408 vitamins C (ascorbic acid) and E (tocopherol), carotenoids, polyphenols such as  
409 anthocyanins, omega-3 and omega-6 fatty acids, and uric acid. Endogenous and  
410 exogenous antioxidants act as “free radical scavengers” by preventing and repairing  
411 damages caused by ROS and RNS (Graves, 2012; Lushchak, 2014; Pham-Huy et al.,  
412 2008).

413 Using different *in vitro* assays such as 2,2-diphenyl-1-picrylhydrazyl (DPPH), 2,20-  
414 azino-bis [3-ethylbenzthiazoline-6-sulphonic acid] (ABTS) and the ferric ion reducing  
415 antioxidant power (FRAP), several studies have shown the antioxidant activity of carob  
416 extracts (Benchikh & Louailèche, 2014; Carbas et al., 2019; Goulas & Georgiou, 2020;  
417 Mekhoukhe et al., 2018; Quiles-Carrillo et al., 2019). Most of these works have studied  
418 different phenolics extraction conditions, and they have directly related the content of  
419 phenolic compounds with the antioxidant activity. Goulas & Hadjisolomou (2019)  
420 studied the effect of triple-step digestion on the stability of phenolics and antioxidant  
421 potency in carob products. They concluded that the carob phenolics are degraded,  
422 released from the food matrix, or attached to other food or digestive juice constituents.

423 Additionally, the critical role of the food matrix was underlined as the food components  
424 had a protective effect against pH changes and enzymatic activities along with digestion.  
425 Other works demonstrated that the antioxidant capacity also depends on the carob fruit  
426 maturity. Benchikh et al. (2014) showed that carob pods have the highest total phenolic,  
427 total flavonoid and ascorbic acid content in the unripe stage. Therefore, the antioxidant  
428 activity decreases significantly throughout the ripening process. Similar results were  
429 obtained by Saci et al. (2019), suggesting that the extract of unripe carob may provide a  
430 substantial source of secondary metabolites, which act as natural antioxidants; moreover,  
431 they have a neuroprotective effect *in vitro*.

432 Several studies revealed a positive correlation between the strong antioxidant activity and  
433 the positive effect on rabbit growth performance (Abu Hafsa et al., 2017),  
434 spermatogenesis (Sadat et al., 2019) and mood disorders (Lakkab et al., 2019).

435 In addition to the antioxidant activity of carob extract due to its strong scavenging effect  
436 on reactive oxygen and free radicals, Rtibi et al. (2015a, 2015b) proved the capacity of  
437 carob pods aqueous extract to inhibit the myeloperoxidase (MPO) activity in a  
438 concentration-dependent manner. The MPO-activity blockage by the carob extracts has,  
439 therefore, the ability to reduce the production of hypochlorous acid (HOCl) from H<sub>2</sub>O<sub>2</sub>.

440 Carob honey is another well studied carob-based product. Its composition depends highly  
441 on the type of flowers utilised by the bees (Wang & Li, 2011). El-Haskoury et al. (2017)  
442 characterised the physicochemical composition and antioxidant activity of carob honey  
443 collected in Moroccos. The antioxidant capacity of the honey samples was correlated with  
444 their biochemical constituents, such as total phenol and flavonoids content. Previously,  
445 El-Haskoury et al. (2015) reported that carob honey has diuretic, natriuretic, and kaliuretic  
446 activity without side effects of hypokalemia observed using furosemide.

### 447 **3.2. Anti-hyperglycaemic and anti-absorptive of glucose properties**

448 An important therapeutic strategy used for diabetes (type 2) regulation and control is the  
449 inhibition of  $\alpha$ -amylase and  $\alpha$ -glucosidase, two key-enzymes in type 2 diabetes mellitus.  
450  $\alpha$ -glucosidase, after its catalysis activity, releases glucose into the blood, which provokes  
451 an increase in its level. Inhibitors of these enzymes delay carbohydrate digestion and  
452 prolong overall carbohydrate digestion time, causing a diminution in the level of glucose  
453 absorption and therefore decreasing the postprandial plasma glucose rise. In this respect,  
454 the work of Custódio et al. (2015) reports the *in vitro* inhibitory activity of water  
455 decoctions of leaves, germ flour, pulp, locust bean gum and stem bark of carob tree on  $\alpha$ -  
456 amylase and  $\alpha$ -glucosidase. Leaves and stem bark decoctions had significant antioxidant  
457 activity and the highest total phenolics content. The major compounds were identified as  
458 gallic acid in the leaves and gentisic acid in the stem bark. Chait et al. (2020) were  
459 pioneers in studying carob anti-diabetic activities during gastrointestinal digestion. These  
460 authors proved the inhibitory effect of carob powder and its digested fractions on  $\alpha$ -  
461 amylase and  $\alpha$ -glucosidase activities. Additionally, they detected ten phenolic acids and  
462 six flavonoids in soluble (free and conjugated) and insoluble fractions of undigested  
463 carob, and, after *in vitro* gastrointestinal digestions, they identified that the most  
464 bioaccessible phenolic acids and flavonoids were gallic acid, chlorogenic acid, (+)-  
465 catechin and rutin. Myricetin and gallic acid were the most abundant metabolites of the  
466 residual fraction of carob phenolics after faecal fermentations. With all this, they  
467 concluded that these carob phenolic compounds might influence glucose metabolism by  
468 inhibiting carbohydrate digestion.

469 Rtibi et al. (2017) developed a study that supports the use of immature (unripe) carob pod  
470 aqueous extract on the reduction of intestinal glucose absorption. They confirmed that the  
471 degree of maturity of carob characterised by a different phytochemical composition,  
472 principally polyphenols and flavonoids, may be responsible for hypoglycemic and

473 hypolipidemic effects. The results proved that intestinal glucose transport inhibition  
474 represents an important component through which immature carob can reduce blood  
475 glucose, probably by the known ability of polyphenols to modify carbohydrate digestion  
476 and absorption. Another possible mechanism may be attributed to fibre in carob pods  
477 because of complex carbohydrate, high molecular weight and viscous, soluble dietary  
478 fibres to retard glucose absorption (Macho-González et al., 2017).

479 As previously referred, carob pods is also a known source of D-pinitol (Tetik et al., 2011).  
480 Inositols and their derivatives, such as inositol-(phospho)glycans have important roles in  
481 physiological processes, principally in modulating the insulin signalling pathway (López-  
482 Gambero et al., 2020). Bañuls et al. (2016) assessed the effects of a 12-week  
483 administration of an oral nutritional inositol-enriched beverage (IEB) on blood glycaemia  
484 (fasting and postprandial states). These authors concluded that IEB daily oral ingestion  
485 for 12 weeks positively influenced insulin resistance and the percentage of glucose  
486 change after main meals and overnight fasting in non-obese prediabetic subjects.  
487 Therefore, inositol from carob extract administration has a potential therapeutic effect in  
488 insulin resistance-related diseases.

### 489 **3.3. Anti-hyperlipidemia properties**

490 Increased postprandial lipemia is reflected with elevated levels of lipids, such as total  
491 cholesterol (TC), low-density lipoprotein cholesterol (LDL), very low-density lipoprotein  
492 cholesterol (VLDL) and triglycerides (TG), which are widely recognised as risk factors  
493 for cardiovascular disease (CVD). High levels of oxidised LDL (OxLDL) is another  
494 relevant risk for oxidative stress and endothelial dysfunction associated with CVD  
495 progression (Chan et al., 2013). In several studies, it has been reported that carob fruit  
496 extract has potential health benefits, including reduction of LDL levels in

497 hypercholesterolaemic patients, beneficial effect on body weight, and improvement of  
498 digestion and lipid utilisation (Abu Hafsa et al., 2017).

499 Macho-González et al. (2018) demonstrated, for the first time, the hypolipidemic  
500 properties of carob fruit extract (CFE) in healthy Wistar rats, modifying postprandial  
501 lipemia by reducing the extents of fat digestion and absorption. These results showed that  
502 the fibre could lower triglycerides, total cholesterol, and low-density lipoprotein  
503 cholesterol. The effect of CFE-enriched meat-consumption on lipemia and lipoprotein  
504 profile in Wistar rats was posteriorly assessed. The authors confirmed the hypoglycemia,  
505 hypoinsulinemia, hypotriglyceridemia and decreased triglyceride-enriched-VLDL in rats  
506 fed with the high-saturated-fat diet containing CFE-enriched meat when compared to a  
507 control group (Macho-González et al., 2019). Additionally, they proposed a mechanism  
508 related to CFE effects on lipoprotein metabolism and insulin signalling (Macho-González  
509 et al., 2020), proving a remarkable and direct relation between antioxidant properties and  
510 postprandial hyperglycemia and hyperlipidemia reduction in healthy animals.

511 Experiments with rabbits showed that insoluble dietary fibre from carob pod could reduce  
512 atherosclerosis development. The results suggested that increased expression of aortic  
513 sirtuin-1 and peroxisome proliferator-activated receptor- $\gamma$  coactivator-1 $\alpha$  may play a key  
514 role in the beneficial effects of carob fibre in dyslipidemia (Valero-Muñoz et al., 2019).

515 All these effects do not seem to be due to only one carob fruit component but rather to  
516 the synergistic action of different constituents. These include polyphenols and flavonoids  
517 of quercetin glycosides, high amounts of insoluble fibre (cellulose and hemicellulose) and  
518 only negligible soluble fibre amounts. Flavonoids may act by making liver cells more  
519 efficient to remove LDL from the blood. For this, flavonoids increase LDL receptor  
520 densities in the liver by binding to apolipoprotein B (El-Beshbishy et al., 2006).

521 Martínez-Rodríguez et al. (2013) assayed a non-extractable-tannates rich carob-fibre (PF-  
522 1®) in a developed milk matrix. After a 4-week interventional study with human  
523 volunteers (400 mL daily consumption of this functional food, containing 20 g of PF-  
524 1®/L), they verified a decrease in triglyceride and total cholesterol levels in all  
525 participants. This preliminary study highlights the probable positive influence of this  
526 functional food on regulating the lipid profile and bowel function in humans.

### 527 **3.4. Gastrointestinal benefits**

528 Gastrointestinal disorders (GID) are responsible for increasing deaths across Europe  
529 annually, with an associated heavy economic burden to the healthcare system (Anderson  
530 et al., 2014). The most common GID include gastritis, peptic ulcers, *Helicobacter pylori*  
531 infection, gastric cancer, and colorectal cancer. In the last years, the gastroprotective  
532 effect of carob pods aqueous extract (CPAE) has been thoroughly studied. Rtibi et al.  
533 (2015b) evaluated the gastroprotective role of the CPAE (15 days) against oxidative stress  
534 induced by acute ethanol exposure in rats. They concluded that CPAE exerts protective  
535 effects against acute ethanol-induced ulceration in the gastric mucosa. This is due to the  
536 antioxidant properties; the CPAE counteracted EtOH-induced gastric lipoperoxidation,  
537 reversed the decrease of sulfhydryl groups (-SH) and hydrogen peroxide levels, and  
538 prevented the depletion of antioxidant enzyme activity of superoxide dismutase (SOD),  
539 catalase (CAT) and glutathione peroxidase (GPx).

540 Other multifactorial gastrointestinal disorders such as constipation and diarrhoea have  
541 been studied in the last years. Rtibi et al. (2016) investigated the putative effects of CPAE  
542 on gastrointestinal transit (GIT), diarrhoea and intestinal epithelium permeability in  
543 healthy rats and mice depending on its degree of maturity. They showed that mature carob  
544 pods' water extract significantly increased the GIT (depending on the doses). In contrast,  
545 the aqueous extract of the immature carob pods induced a significant decrease in the GIT.

546 They suggested that there is a difference in the chemical composition of both extracts.  
547 The extract of mature carob pods has high fibre and sugar contents; these compounds are  
548 absent or present in low quantity in immature carob pods. It was established that fibres  
549 and sugars in high concentrations cause the acceleration of the GIT process (Mudgil &  
550 Barak, 2013). The high total tannins content present in the immature carob pods may be  
551 responsible for inhibiting GIT and diarrhoea. Concerning the intestinal epithelium  
552 permeability, the aqueous extract of immature carob pods causes an inhibition of the  
553 secretion, implying a transit slowdown. In contrast, the authors showed that the aqueous  
554 extract of mature carob pods causes an increase in the intestinal secretion, facilitating the  
555 GIT.

556 The protective effect of carob in ulcerative colitis, another gastrointestinal diseases  
557 related to inflammation and oxidative stress, has also been studied (Aboura et al., 2017;  
558 Rtibi et al., 2016). The analysis of myeloperoxidase activity, cytokines and oxidative  
559 damage after the dextran sulfate sodium-induced sub-acute experimental ulcerative colitis  
560 in the rat model, with and without the administration of carob extract, determined that the  
561 high polyphenols content of carob fruit prevented colonic shortening and reduced the  
562 colonic severity lesions and biochemical alterations.

#### 563 **4. Food applications**

564 Great challenges for the food industry are the clear demonstration of the health benefits  
565 of natural ingredients sources before they can be successfully incorporated into functional  
566 food products with regulatory compliance and consumer acceptance (McClements &  
567 Xiao, 2014). Several studies in recent years have demonstrated the versatility and  
568 functionality of different carob products used in the production of functional foods. The  
569 processing of whole carob fruit to obtain different carob products significantly affects  
570 physicochemical and functional properties. The carob products can be used as natural

571 ingredients in the food industry due to flavouring or colouring effects and improved final  
572 products' characteristics. Some examples of foodstuffs with the addition of carob products  
573 are presented in Table 4. The enhanced properties of the final products are highlighted.  
574 The processing conditions of carob pods to obtain syrup and powder defines the  
575 functional properties required by the food industry. Carob syrup (60, 70 or 80 °Brix) were  
576 characterised by important reducing sugars content, dark colour and functional properties  
577 with high antioxidant activity and emulsifying capacity. These improved properties can  
578 be attained from non-enzymatic browning reactions occurring during juice boiling.  
579 Microwave drying at different power levels (100, 300 and 600 W) is useful for  
580 dehydrating the by-product obtained after juice extraction and producing carob powders  
581 with high dietary fibres, brown colour, and water/oil retention capacity (Tounsi et al.,  
582 2017). Different degrees of carob flour roasting can also affect the physicochemical  
583 characteristics and acceptability of different bakery products. In cakes and cookies, the  
584 incorporation of carob flour increased the viscosity of cake batters. It increased the solid  
585 elastic-like behaviour of the cookie doughs, indicating a more robust interaction of the  
586 ingredients. The addition of highly roasted carob flour only reduced the acceptability of  
587 cakes. Simultaneously, in the cookies, there was a decline in the acceptability of all carob  
588 flour cookies (Román et al., 2017). Srour et al. (2016) also proved that beverages  
589 formulated with roasted carob powder had higher ratings for residue levels, colour, odour  
590 (caramel, mocha, and roasted coffee), flavour (mocha and roasted coffee), viscosity,  
591 mouthfeel, and bitter aftertaste.

592 Furthermore, it is important to understand the physical and chemical properties of carob  
593 products and their interdependence to use them efficiently as a functional food ingredient.  
594 For example, when Benković et al. (2017) studied physicochemical properties of carob  
595 flours produced with and without seeds, they concluded that carob flours containing seeds



596 had a higher cohesion index and higher cake strength values than flours without seeds.  
597 Additionally, the extraction efficiency of polyphenols and flavonoids and the antioxidant  
598 capacity depended on the cohesion and cake formation ability of flours. Regarding the  
599 carbohydrates, samples without carob seeds established higher total sugar and soluble  
600 polysaccharides content.

601 When Fidan et al. (2019) replaced wheat flour with 50% carob flour as a functional  
602 ingredient in sponge cakes resulted in an increase in dietary fibre (from 2.45 to 18.28  
603 g/100 g dry weight), protein (from 8.44 to 23.93 g/100 g dry weight) and carbohydrate  
604 content (from  $65.40 \pm 5.20$  to  $86.10 \pm 2.70$  g/100 g dry weight). The cakes with carob flour  
605 showed differences in cell size and uniformity and sweetness and crumb tenderness,  
606 confirming that the replacement of ingredients positively influenced sensory  
607 characteristics. These properties add nutritional and biological values to traditional  
608 culinary products, also depending on the remaining ingredients. Papaefstathiou &  
609 Agapiou (2018) studied the nutritional composition of twenty traditional carob food  
610 products, comparing results with the pulp of Cypriot carob cultivars. They concluded that  
611 despite carob fruit and carob powders could be recommended to be included in humans'  
612 daily diet as they contain valuable nutrients and low fat while having a sweet taste, carob  
613 products only partially satisfied healthy and nutritional claims: 60% in terms of fat, 25%  
614 in terms of dietary fibres, and 80% in terms of minerals.

615 Another important benefit of carob product is that they can be used in the bakery industry  
616 as gluten-free flour. Several authors investigated the effects of incorporating carob flour  
617 in several gluten-free cakes, bread, and snacks. Martin-Diana et al. (2017) developed a  
618 gluten-free cracker snack through the inclusion of carob by-products (germ and seed  
619 peel). They concluded that germ and seed peel from carob processing by-products  
620 provided protein and antioxidant activity to the formulated food products. Indeed, by the

621 composition of both raw materials, the germ and peel concentration had a significant  
622 linear effect on the protein and carbohydrate (fibres) contents, respectively. Regarding  
623 the phenolic content, the inclusion of germ increased it, reaching the highest values at  
624 5.44% germ and no peel. The phenolics corresponded to proanthocyanidin, gallotannins,  
625 tannins, catechins and flavonols (Albertos et al., 2015), and these polyphenols would be  
626 responsible for the antioxidant activity of the product. In addition to the nutritional profile,  
627 the physicochemical parameters and sensorial analysis were determined. In the first case,  
628 they observed that at maximum concentrations of germ (17.6%) and concentrations close  
629 to 6.5% for peel, the product absorbed and retained the maximum water proportion water.  
630 This behaviour could be attributed to the presence of remaining gums (galactomannan)  
631 from the endosperm. The water activity ( $a_w$ ) also was analyzed since it is an indicator of  
632 quality for snacks.  $a_w$  decreased with increasing germ and peel concentrations. Also,  
633 firmness and hardness increased linearly with the increment of germ on the formulation.  
634 Regarding the sensory analysis, the cracker with germ and peel ranked the lowest in  
635 acceptability. They affected the general acceptability of the final product, especially the  
636 germ, probably because of the high content of tannins and catechins, which provide  
637 important bitter and astringent flavour. Colour differences among crackers may have also  
638 affected the general acceptability.

639 In another work, Berk et al. (2017) investigated the effects of partial replacement of rice  
640 flour by carob bean flour at different concentrations (10%, 20%, and 30%) in gluten-free  
641 cakes. First, the specific gravity was determined, observing that the increased carob bean  
642 flour amount in the formulation led to the thicker batter, which might be due to increased  
643 fibre and sugar content. Increasing fibre content might obstruct mixing efficiency and  
644 aeration of gas bubbles. Then, the specific volume of cakes was studied. Although it did  
645 not create a significant difference, 20% carob bean flour added samples had relatively

646 higher volume than 10% and 30% carob bean flour containing cakes, respectively.  
647 Textural analysis of cakes was evaluated in terms of hardness. Increasing carob bean flour  
648 concentration from 10% to 20% and 30% created a significant difference in hardness.  
649 Since air incorporation in cake batter during mixing is one of the factors that affect the  
650 hardness of cakes, increasing carob bean flour concentration in formulation might make  
651 incorporation of air in batter difficult with increasing viscosity. This might be due to not  
652 only increase in fibre content but also soluble components in the formulation. Due to the  
653 high-water holding capacity of fibre, the viscosity of cake batter tended to increase with  
654 concentration.

655 Claudia Arribas et al. (2020) determined the content of phenols in dry (uncooked) and  
656 cooked experimental samples based on different proportions of rice/bean and  
657 supplemented with whole carob flour (WCF), as well as in a commercial rice-based pasta  
658 whose phytochemical content has not been previously studied. The pasta quality was  
659 assessed by determining the texture and colour properties. The supplementation of the  
660 different rice/bean samples with 10% WCF increases the content of phenolic compounds,  
661 both in the uncooked and the cooked samples. The antioxidant activity was linked to the  
662 presence of phenols supplied by the bean and WCF flours and other antioxidant  
663 compounds such as the Maillard reaction products formed during the pasta-making drying  
664 step. As mentioned above for the phenolic compounds, the presence of WCF increased  
665 the antioxidant activity in comparison to the same formulations without this legume. The  
666 colour parameters of both the cooked and uncooked fettuccine showed significant  
667 differences between the samples with or without WCF. These differences can be linked  
668 to the brown colour of the raw WCF and the processing conditions, both in the making  
669 and the cooking processes. The supplementation of fettuccine with 10% WCF decreased  
670 the hardness compared to the same pasta without WCF, probably due to the increase in

671 dietary fibre content, which may have led to crashes or breaks inside the fettuccine strand,  
672 thus weakening the pasta structure. The improvement of the texture parameters observed  
673 could meet the expectation of consumers of GF products.

674 Gluten-free bread from rice flour substituted with carob flour and resistant starch (RS)  
675 was investigated (K. Tsatsaragkou et al., 2014). Carob flour addition increases the water  
676 absorption of the dough. Increased amounts of carob flour caused a reduction in moisture  
677 values, for the same water content, due to the increased water holding capacity of carob  
678 flour, ascribed to both protein (caroubin) and dietary fibres (locust bean gum) content.  
679 Carob and water content did not significantly affect porosity, surface porosity and  
680 elasticity values. Porosity values were greater than those found for bread without carob  
681 flour, whereas elasticity values were lower.

682 In several works (Arribas et al., 2017; Arribas et al., 2019a, 2019b) studied the effect of  
683 extrusion on nutritional components, the bioactive compounds content and textural  
684 analysis of various gluten-free formulations based on carob fruit, pea and rice flour  
685 blends. The extruded formulations containing whole carob fruit can be considered a good  
686 source of protein, dietary fibre, and phenolic compounds (anthocyanins, flavonols,  
687 tartaric esters, and total phenols). The antioxidant activity and the different groups of  
688 phenols showed a positive correlation in the extrudates. On the other hand, a higher  
689 amount of carob flour caused a lower preferred colour by the panellists. This could be  
690 related to the brown colour of the products formulated with carob flour, leading to a slight  
691 rejection of these products. The analysis of the different aroma and taste parameters tested  
692 scores did not show any correlation with the amount of legumes in the extrudates and no  
693 unpleasant odours or flavours were detected. In conclusion, the addition of 5% carob fruit  
694 flour did not significantly affect the overall quality of these extrudates, and therefore these  
695 novel extrudates would be well accepted by consumers.

696 (Tsatsaragkou et al., 2017) determined the influence of the particle size of carob seed  
697 flour on the quality of fresh and stored gluten-free rice-based bread. Bread made with  
698 carob flour showed a lower specific volume than commercial mixtures. Among the carob  
699 breads, the incorporation of coarser (FA) and finer (FD) fraction led to the production of  
700 breads with increased specific volume. Their lower amount of protein than the other  
701 fractions and the higher amount of dietary fibres (polysaccharides from the endosperm of  
702 the carob seed) might improve the specific volume. On the other hand, the addition of  
703 carob flour to produce gluten-free rice breads significantly modified the crumb colour and  
704 presented lower firming rate; thus, the presence of carob flour had an anti-staling effect  
705 that was independent on the particle size distribution.

706 Given the use of seed germ flour for food applications, carob seed was characterised by  
707 high protein content (25.7%), while mannose and galactose were the dominating  
708 monosaccharides. Moreover, the isolated galactomannan from carob seeds demonstrated  
709 good swelling properties, *i.e.*, 30.1 ml per g sample and oil-holding capacity (27.9 g/g  
710 sample) (Fidan et al., 2020). Attenuated total reflectance-Fourier transform infrared  
711 spectroscopy (ATR-FTIR) analyses detected a high proportion of  $\beta$ -sheet conformation  
712 for *C. siliqua* germ proteins, suggesting that carob germ flour could be technologically  
713 best suited for use in baked and cooked food formulations (Mamone et al., 2019). Turfani  
714 et al. (2017) proved that blending wheat flour with carob seed flour at 5-6% or 10-12%  
715 modified the dough's technological properties and bread characteristics. Carob seed flours  
716 increased dough tenacity while reducing extensibility. This is probably related to the  
717 tenacity of the hydrated carob fibres and the viscoelastic properties of the germ caroubins.  
718 Germ caroubins, like gluten, can form fibrils and sheets and contain large polymeric  
719 proteins that are cross-linked by disulfide bonds. On the other hand, carob seed flours  
720 increased water absorption up to about 40%. This behaviour is related to the fact that the

721 great number of hydroxyl groups and the carob soluble fibre structure allow more  
722 hydrogen bonding with water than gluten and starch (Barak & Mudgil, 2014). Moreover,  
723 in the case of carob seed germ flour, it is reported that caroubins are more hydrophilic  
724 than gluten (Smith et al., 2008). Regarding the specific volume, its increase or decrease  
725 depended on the amount and type of carob flour. When baking blends at 5% level carob  
726 flour substitution, it did not present differences with respect to the wheat flour control.  
727 However, a proportion of 10% or more of substitution with carob seed germ flour reduced  
728 the loaf volume. In contrast, refined carob seed flour produced a statistically significant  
729 increase of loaf volume and raw carob seed flour a slight increase that was not significant.  
730 This is attributed to reduced extensibility and weakening of the gluten network due to  
731 dilution, reduced hydration, and interaction with fibres and non-gluten proteins, reducing  
732 the gas-retention ability. Carob flours enriched bread with soluble dietary fibres, insoluble  
733 dietary fibres, phenolic compounds and lignans, thus enhancing its antioxidant properties.  
734 The addition of raw carob seed and carob germ flours increased polyphenols in bread,  
735 especially the free ones. Two lignans, secoisolariciresinol and isolariciresinol, were found  
736 in wheat bread.

737 In some cases, reactive 1,2-dicarbonyl compounds (DCs) are generated from  
738 carbohydrates during food processing and storage (Hellwig et al., 2018). They are formed  
739 mainly by dehydration and redox reactions. They substantially impact on food palatability  
740 because they have an essential role in aroma and colour formation. They can also be  
741 formed under physiological conditions, covalently modifying proteins *in vivo* and impair  
742 their function. However, they are precursors of advanced glycation end products (AGEs),  
743 and cytotoxic effects of several DCs have been reported. The most abundant DCs in food  
744 are 3-deoxyglucosone, 3-deoxygalactosone, and glucosone, predominating over  
745 methylglyoxal, glyoxal, and 3,4-dideoxyglucosone-3-ene. 3-Deoxyosones are precursors

746 of furan derivatives such as furfural and hydroxymethylfurfural (HMF). HMF is a  
747 prominent indicator of thermal treatment of carob flour (Vitali Čepo et al., 2014) and  
748 syrup (Sengül et al., 2007). Even so, it is essential to quantify these compounds and to  
749 understand their physiological effects.

750 Finally, lipid oxidations can affect the quality characteristics and safety of lipids foods  
751 and emulsions, losing their nutritional value. Goulas & Georgiou (2020) proved the  
752 potential of carob extracts to prevent lipid oxidation in food systems such as emulsions,  
753 sunflower oil, and cooked comminuted pork. Thus, the food industry must foster natural,  
754 safe, and low-cost antioxidant compounds to substitute synthetic additives such as  
755 butylated hydroxyanisole and butylated hydroxytoluene.

## 756 **5. Data Collection**

757 Our research, based on a framework for scoping review, included a thorough search of  
758 published literature in the last 15 years to collect a significant amount of information  
759 related to carob and its properties. The main keywords used were carob, chemical  
760 composition, pulp, seed, health, biological activity, phenolics, locust bean gum, pinitol,  
761 amino acids, sugars, fibres, minerals, functional food. The databases searched were  
762 Scopus and Web of Science. The quality assessments of the papers were not performed.

## 763 **6. Conclusion**

765 The current trends of a healthy lifestyle for health promotion and disease prevention have  
766 led the food industry to seek innovative health-beneficial food products based on natural  
767 sources. The high dietary fibre content and bioactive compounds of carob fruit and its  
768 products (powder, flour, and syrup), and their beneficial effects on gastrointestinal  
769 diseases, diabetes, hyperlipidemia, inflammation, and oxidative stress make carob  
770 products novel food ingredients, which have potential to be used in the development of a  
771 wide variety of health-beneficial food products. The incorporation of carob fruit and its

772 derived products in food formulations enhance nutritional and functional value and  
773 improve technological functionality by imparting beneficial rheological properties and  
774 extending the shelf life of the final products. At present, carob flour or powder production  
775 is only done after the pulp separation. However, it has been demonstrated that the seed  
776 presence improves the cohesion index, food product strength values, the extraction  
777 efficiency of polyphenols and flavonoids, and the antioxidant capacity of the products.  
778 Consequently, it is crucial to understand the physical and chemical properties of carob  
779 products and their interdependence to use them efficiently as functional food ingredients.

780 *Ethics Statement*

781 None.

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788 *CRedit author statement*

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793 *Declaration of Competing Interest*

794 The authors have declared no conflict of interest.



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## Figure Legends

Figure 1. a) Carob tree (*Ceratonia siliqua* L.), b) Immature and mature carob pod, c) Carob pod constituents (Tous & Antoni, 2013).

Figure 2. Main industrial uses of carob fruit (Durazzo et al., 2014; Ersan et al., 2020; Tounsi et al., 2017; Yousif & Alghzawi, 2000).

**Table 1.** World average carob statistics from 2015 to 2018 (FAOSTAT, 2020).

	<b>Portugal</b>	<b>Italy</b>	<b>Morocco</b>	<b>Turkey</b>	<b>Greece</b>	<b>Cyprus</b>	<b>Algeria</b>	<b>Spain</b>	<b>World</b>
<b>Area harvested (ha)</b>	13,427	5,600	10,224	3,099	2,410	1,004	808	2,292	41,593
<b>Yield (hg/ha)</b>	29,393	56,385	21,532	45,776	59,377	71,571	45,762	8,382	32,839
<b>Production (tonnes)</b>	39,387	31,577	22,013	14,195	12,819	7,179	3,701	1,916	136,613
<b>Production (%)</b>	28.83	23.11	16.11	10.39	9.38	5.25	2.71	1.40	100

**Table 2.** Chemical composition of carob powder

<b>Component</b>	<b>Proportion (%)</b>
Moisture	6.3 to 7.6
Protein	1.7 to 5.90
Ash	2.3 to 3.2
Fat	0.2 to 4.4
Total dietary fibre	11.7 to 47
Starch	0.1
Total Carbohydrates	42 to 86
Fructose	2 to 7.4
Glucose	3 to 7.3
Sucrose	15 to 34
D-Pinitol	5.5

Source: (Faik A. Ayaz et al., 2009; Carbas et al., 2019; Özcan et al., 2009; Petkova et al., 2017).

**Table 3.** Carob chemical constituents and their biological properties associated with health benefits.

<b>Chemical Constituents</b>	<b>Biological Properties/Health benefits/Disease</b>	<b>Carob Part/Fraction</b>	<b>Reference</b>
Polyphenols/Gallic acid and Gallotannins/Pyrogallol/Chlorogenic acid, (+)-Catechin, Rutin, Myricetin.	Glycemic control/Neurogenerative diseases/Antioxidant, anti-inflammatory and antitumoral activities /Abdominal obesity control/Antibacterial activity	Carob pulp powder	Chait et al., 2020; Fidan et al., 2018; Martínez-Villaluenga et al., 2018; Papakonstantinou et al., 2018; Rico et al., 2019; Rodríguez-Solana, Coelho, et al., 2019; Rtibi et al., 2015a
Polyphenols/Epigallocatechin gallate, Epigallocatechin, Catechin, Syringic acid, Quercetin glycoside, Gallic acid, Catechin gallate, Cinnamic acid	Antioxidant, Antibacterial, Antifungal activities/Cytotoxicity effects/Hepatoprotective effects	Carob pulp extract	Abulyazid et al., 2017; Dhaouadi et al., 2014
Insoluble dietary fibre	Enhanced lipoprotein metabolism, lowers total and LDL cholesterol/Hypoglycemic effect	Carob pulp	Macho-González et al., 2017, 2019, 2020; Valero-Muñoz et al., 2019
D-pinitol	Anti-diabetic activity/Anticancer/Hepatoprotective	Carob pulp	Bañuls et al., 2016; Christou et al., 2019; López-Sánchez et al., 2018
Polyphenols, Flavonoids, and condensed tannins/Quercetin and Apigenin derivatives/Tannic acid	Antioxidant and Antihypertensive activity/Abdominal obesity control/Anxiolytic and Antidepressant effect	Carob seed peel	Lakkab et al., 2019; Rico et al., 2019
Flavonoids and tannins/Fibre and sugar/pyrogallol, catechin, gallic acid, tannic acid, chlorogenic acid, and epicatechin.	Antibacterial activity/ Anti-ulcer and Gastroprotective effect/ Laxative and Anti-diarrheal Activities	Carob pod	Abu Hafsa et al., 2017; Rtibi et al., 2015b, 2016
LBG/galactomannan	Gastrointestinal effects	Seed endosperm	(Xie et al., 2020)



**Table 4.** Carob-based functional foods and main benefits ascribed.

Functional food	Positive effects	References
Wafer cream with carob pod and chicory root powder	<ul style="list-style-type: none"> <li>• Low caffeine content</li> <li>• High amount of sugar</li> <li>• High antioxidant activity</li> <li>• Positive effect on physicochemical, rheological, and sensory properties</li> </ul>	Habibzadeh & Seyedain Ardabili, 2019
Rice-based extruded snacks-like fortified with pea, bean and carob fruit	<ul style="list-style-type: none"> <li>• High phenolic compounds content</li> <li>• High antioxidant activity</li> <li>• Improved textural attributes</li> </ul>	Arribas et al., 2019b, 2019a
Muffin with carob powder	<ul style="list-style-type: none"> <li>• High water activity</li> <li>• High total phenolics content</li> <li>• High antioxidant activity</li> <li>• High content of phytosterols and genistein</li> <li>• Increased browning index and FAST index</li> </ul>	Červenka et al., 2019; Pawłowska et al., 2018
Bread with carob flour (pods and germ flours)	<ul style="list-style-type: none"> <li>• Improved texture and sensory properties</li> <li>• Antimicrobial activity</li> <li>• High total phenolics content</li> <li>• High antioxidant activity</li> <li>• High gluten-aggregation</li> <li>• Improved bread quality</li> </ul>	Hoehnel et al., 2019; Rico et al., 2018; Šoronja-Simović et al., 2016
Sponge cakes enriched with carob flour and carob syrup	<ul style="list-style-type: none"> <li>• Higher level of dietary fibre, protein, and carbohydrate contents</li> <li>• Higher total moisture content</li> <li>• Positive influence in sensory characteristics</li> </ul>	Fidan et al., 2019
Carob spread	<ul style="list-style-type: none"> <li>• Good colour and textural characteristics</li> <li>• Excellent source of essential minerals</li> <li>• High total phenolic content</li> <li>• Low quantities of Hydroxymetylfurfural (HMF)</li> </ul>	Aydın & Özdemir, 2017
Pasta enriched with carob flour	<ul style="list-style-type: none"> <li>• Good colour, firmness and hardness</li> <li>• high antioxidant activity and sucrose.</li> <li>• High total phenols content</li> <li>• High Glycemic index</li> </ul>	Arribas et al., 2020; Biernacka et al., 2017; Sęczyk et al., 2016
Sesame paste enriched with carob syrup	<ul style="list-style-type: none"> <li>• Improved emulsion stability</li> <li>• High nutritional values</li> </ul>	Tounsi et al., 2019
Low lactose yoghurt with carob flour	<ul style="list-style-type: none"> <li>• High fibre content</li> <li>• High sweetness content</li> </ul>	Moreira et al., 2017