


Article

Coffee Roasting, Blending, and Grinding: Nutritional, Sensorial and Sustainable Aspects

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Abstract: The objective of this work was to evaluate the influence of roasting, blending, and grinding on the nutritional, sensory and sustainable aspects of coffee. To achieve this, a systematic review of the literature was performed. The database for the selection of relevant papers was the Portal de Periódicos da Capes, with remote access via CAFE. For the elaboration of the research, a chronological criterion with period restriction was used, considering the period between 2008 and 2022, to access all possible works related to the theme of this work. The following terms were used: blending; grinding; coffee; nutritional; sensory; sustainability; and roasting. To filter the searches, the association of these terms was also used by means of links and word associations. In the terminology, the Boolean operator “AND” was used to interconnect the terms used. The roasting degree, grinding, and the amount of each coffee species impact the nutritional and sensorial aspects of coffee, while the determination of each blend influences the sustainability of the environmental, economic and social aspects of the coffee production chain.

Keywords: sustainability; coffee production; quality



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1. Introduction

Brazil is the main producer of coffee worldwide, producing 63,400 thousand 60 kg bags in 2020, as well being the main exporter of this product, exporting 40,511 thousand 60 kg bags in 2020 [1]. In addition, it is the second largest consumer of coffee, at above 1.2 million of tons, with a per capita consumption of 5.96 kg of green coffee or 4.77 kg of roasted coffee [2]. Coffee comes from plants belonging to the *Coffea* genus, including two main species used commercially, *Coffea arabica* L. and *Coffea canephora*, known as Arabica and Robusta coffee, respectively.

Arabica coffee comprises 70.0% of Brazilian production, with an estimated production of 2.29 million of tons in 2019, whereas 979,800 tons of robusta coffee was produced in the same year [3]. Minas Gerais, a state in Brazil, is the main producer, and provides 50.8% of the total production in Brazil, mostly consisting of Arabica coffee. Espírito Santo, another state in Brazil, is the second largest producer, mainly cultivating robusta coffee, providing around 76.5% of Brazil's output of this coffee type [3].

These two species differ from each other regarding the physical and sensorial characteristics of the fruit, the propagation process, and the flowering period, among other aspects. Because of its higher popularity, Arabica coffee is more often grown worldwide, because it provides a higher sensorial quality when compared to Robusta coffee [4]. On the other hand, some characteristics of Robusta coffee, such as its lower susceptibility to diseases, higher productivity, adaptation to lower altitudes below 400 m, and increased tolerance to temperature, ranging from 22 and 26 °C, have increased its market share [5]. In addition, Robusta coffee produces a drink with a fuller body, which is an important sensorial characteristic for many consumers. Thus, blends (mixtures) between these two species are often produced [6]. Blends can be accomplished using different coffee varieties

within the same species; however, this is not common when compared to blends between Arabica and Robusta coffees. Thus, this article will focus on blends between Arabica and Robusta coffee.

Coffee blends have the goal of exploiting the sensorial characteristics of different coffees, merging them to develop different flavors and aromas in the final product, in order to cater to a specific market. Furthermore, in addition to the sensorial aspects of the blends, the sustainability of the business and social aspects is important.

Adding Robusta coffee at a high proportion is not widely accepted by consumers [7], because despite its aforementioned full-bodied nature, it leads to a bitter taste, directly proportional to the amount of Robusta coffee [4]. Additionally, blends can be made with different cultivars from the same species, with the same objective as stated before, i.e., to reach different markets.

There are different approaches to formulating coffee blends. Traditionally, this can be accomplished with raw coffee beans or roasted coffee. In the first case, prior to roasting, the proportions of Arabica and Robusta coffee can be established and then submitted to the roasting procedure. However, due to the differences between these two species (i.e., form, size, composition), roasting a mixture of coffees may lead to different roast degrees of each bean, providing under- or overroasting of the batch. Thus, the second approach, using mixture of roasted coffees, is often applied. After roasting Arabica coffee and Robusta coffee in a separate manner, these batches are submitted to grinding, and then blends are made according to the proportion of Arabica and Robusta coffee.

In contrast to the cited techniques, a different approach to brew manufacturing was presented by [8]. They used a reversed method, grinding first, then roasting. The particle size of the coffee was 21.0% lower and the amount of trigonelline was higher for the reversed method when compared to the conventional method. Also, the profiles of the two samples were slightly different [8]. The authors indicated that the reversed method requires a lower amount of energy, indicating a more sustainable approach. Nevertheless, it is known that both roasting and grinding, regardless of the sequence, have an impact on the cup quality of coffee.

Throughout roasting, different chemical components are formed, contributing to the final aroma of the coffee drink [9]. Acids, lactones and other phenolic-derived components are formed after the roasting of the green grain through the degradation of chlorogenic acids. This trend influences the aroma and flavor of the coffee drink, along with the astringency of the drink and the final acidity [10].

Comminution, or simply the grinding procedure, may provide different particle sizes according to the market's needs [11]. This process seeks to increase the specific surface area of the product, enabling an increase in the amount of compounds extracted [12]. Grinding directly impacts water sorption due to the increment of interactions between coffee and the environment [13]. Ref. [14] stated that smaller particles of coffee lead to lower values of the equilibrium moisture content. Ref. [15] reported that grinding ruptures the coffee tissues and cells, releasing the volatile compounds that contribute to the coffee aroma.

Taking this into consideration, this work aims to review recent studies regarding the nutritional, sensorial and sustainable aspects of coffee and how they are affected by roasting, blending and grinding.

2. Materials and Methods

This research work is characterized by being a systematic review of the literature, in which, according to [16], the databases consulted, the search forms used in these databases, the parameters of the selection process of scientific articles, as well as the criteria for inclusion and exclusion of articles and the process of analysis of each article are indicated.

The database used for the selection of relevant papers was the Portal de Periódicos da Capes, with remote access via I. For the elaboration of this research work, a chronological criterion with period restriction was used, considering the period between 2013 and 2022, to access all possible works related to the theme of this work.

The terminological criterion was also used, which aimed to find articles by means of terms and keywords. The following terms were used: blending; grinding; coffee; nutritional; sensorial; sustainability; and roasting. To filter the searches, the association between these terms was also used by means of links and word associations. In the terminology, the Boolean operator “AND” was used to interconnect the terms used. Thus, the following searches were performed: blending AND grinding; blending AND coffee; blending AND nutritional; blending AND sensorial; blending AND sustainability; blending AND roasting; grinding AND coffee; grinding AND nutritional; grinding AND sensorial; grinding AND sustainability; grinding AND roasting; coffee AND nutritional; coffee AND sensorial; coffee AND sustainability; coffee AND roasting; nutritional AND sensorial; nutritional AND sustainability; nutritional AND roasting; sensorial AND sustainability; sensorial AND roasting; sustainability AND roasting.

In the choice of articles for this work, we analyzed which papers would be more pertinent to the subject in question, excluding articles that were not relevant according to their title and considering the pre-established publication period mentioned above. The articles used further were those that contained the terms sustainability, coffee, grinding, blending, and roasting in their title. Then, the abstract of the selected articles was read to classify them as pertinent or not. The articles were suitable when the abstract indicated that the paper presented results related to the sensory and/or economic and social aspects of coffee production. The non-pertinent articles were discarded, and the selected papers were read completely and used in the present study.

3. Results and Discussion

3.1. Coffee Blends: Development and Innovative Aspects

Coffee blends have the objective of exploiting the sensorial potential of different species, or different varieties within the same species. Robusta coffee has higher productivity while Arabica coffee possess a higher price; thus, blends between these two species provide volume, price stability and different sensorial attributes, because of their different chemical compositions (Table 1).

Table 1. Chemical composition of green Arabica and Robusta coffee.

Composition	Dry Matter Range (%)	
	Arabica	Robusta
Kahweol	0.7–1.1	NA
Caffeine	0.6–1.5	2.2–2.7
Chlorogenic acids	6.2–7.9	7.4–11.2
Sucrose and reducing sugars	5.3–9.3	3.7–7.1
Total free amino acids	0.4–2.4	0.8–0.9
Strecker-active	0.1–0.5	0.2–0.3
Araban	9.0–13.0	6.0–8.0
Reserve Mannane	25.0–30.0	19.0–22.0
Reserve Galactan	4.0–6.0	10.0–14.0
Other polysaccharides	8.0–10.0	8.0–10.0
Triglycerides	10.0–14.0	8.0–10.0
Proteins	12.0	12.0
Trigonelline	1.0	1.0
Other lipids	2.0	2.0
Other acids	2.0	2.0
Ash	4.0	4.0
Totals *	90.0–114.0	86.0–107.0

* Totals of the lower and upper values reflect the scope of the variations of 100% of the dry matter in particular coffees. Source: [17].

Sustainability, in recent years, has become an essential aspect to observe during the production chain of products, such as coffee. This trend is related to consumers’ requirement regarding how coffee is produced, from an environmental, social and sustainability

perspective. Thus, coffee production nowadays must declare the farm location, organic farming practices, whether native forests and biodiversity were preserved, which coffee varieties and fertilization methods were used, the harvest and post-harvest procedures, and the blend composition, among others, depending upon the consumer.

Consumers are displaying increasing interest regarding the aroma, flavor, and color of roasted and ground coffee, evaluating its sensorial characteristics, leading the coffee industry to seek to achieve higher quality of its products by means of acceptability tests, using sensorial analysis, which depends upon the physical and chemical characteristics of the product [18]. To cater to different consumers, chemical and sensorial analysis of these blends should be performed. Sensorial analysis permits one to analyze in a scientific and objective manner the features that influence the acceptability of a food by consumers, utilizing the senses of an integrated team, trained or not, to identify different organoleptic characteristics of the product. This analysis evaluates the intensity of the sensorial attributes of several products, allowing researchers to obtain a complete description of the differences between samples, facilitating the modification of the characteristics of the studied product to address consumer demands [19]. Several investigations regarding coffee blends have been performed in recent years, regarding chemical composition, sensorial analysis, and other important aspects (Table 2).

Table 2. Articles encountered regarding coffee blends and their contributions regarding nutritional, sensorial and sustainable aspects.

Coffee Types Blended	Blending Ration	Main Findings	Reference
Arabica and Robusta (A/R)	(A/R): (100, 0; 90, 10; 80, 20; 60, 40; 40/60; 20/80; 0/100%)	Blends with up to 40% Robusta coffee were accepted by the consumers	[20]
Special Blend (SB) and Market Blend (MB)	SB coffee: 100% Arabica with different roasting degrees MB coffee: 5 coffee brands, 4 were 100% arabica and 1 with some Robusta	Decrease in body fat and increases in energy and nutrient intake were more pronounced with the consumption of SB coffee	[21]
Arabica and Robusta (A/R)	(A/R): (0/100; 15/85; 20/80; 25/75; 35/65%)	Coffee brews prepared from blended coffee beans were well accepted by sensory panelists	[22]
Arabica, Robusta and defective coffee	100% Arabica; 50% of Arabica and 50% defective coffee; 100% Robusta; 50% of Robusta and 50% of defective coffee	The coffee species used had more relevance for differentiating the sensory characteristics of the brews than the addition of defective coffee	[23]
Arabica and Robusta (A/R)	(A/R): (100:0; 50:50; 25:75; 0:100%)	All samples containing Arabica coffee presented amplification for real-time PCR	[24]

Source: the authors.

Ref. [25] analyzed different coffee blends comprising Arabica and Robusta coffees, regarding the acrylamide content, which is probably carcinogenic to humans. They concluded that the acrylamide content increases when the percentage of Robusta coffee within the blend increases.

Ref. [20] verified the acceptance of 112 coffee consumers regarding different blends, containing 0% (100% Arabica), 10%, 20%, 40%, 60%, 80% and 100% Robusta coffee. Beverages containing up to 40% Robusta coffee were accepted by the consumers, whilst beverages with up to 20% Robusta coffee maintained the desired sensory characteristics, such as high intensities of a chocolate aroma, a coffee aroma and flavor, a sweet aroma, and a sweet taste.

Ref. [21] studied the effect of different coffee blend compositions on the body weight, food intake, satiety markers and DNA (deoxyribonucleic acid) integrity of 84 healthy subjects. It was reported that 100% Arabica coffee had a more pronounced effect on body fat, energy and nutrient intakes when compared to coffees containing Robusta beans.

Ref. [26] investigated soaking Robusta beans in solutions of glucose, fructose and sucrose, concluding that this procedure impacted aroma generation during roasting, leading

to altered levels of pyrazines, furans, ketones, organic acids and heterocyclic nitrogen-containing compounds.

Ref. [27] used a pretreatment of Robusta coffee with acetic acid, which provided a closer aroma profile to Arabica, permitting a higher proportion of Robusta coffee in the blends, from 20% up to 80%.

Ref. [22] indicated that a spray-dried coffee blend containing green Arabica coffee at a rate of up to 35% with roasted Robusta beans was well accepted by sensory panelists, along with possessing a greater total phenolic content and antioxidant activity, when compared to 100% Robusta, 15:85 Arabica/Robusta, 20:80 Arabica/Robusta and 25:75 Arabica/Robusta.

Ref. [23] researched the overall acceptance of 100% Arabica coffee, 50% Arabica coffee and 50% steamed defective coffee, 100% Robusta coffee, and 50% Robusta coffee and 50% steamed defective coffee. It was concluded that, despite the differences in caffeine, trigonelline, melanoidin, and total soluble solid content, pH, and acidity, the addition of 50% defective steam-treated *C. canephora* coffee to *C. arabica* and *C. canephora* did not generate different sensory attributes in the blends from those used to describe pure coffee brews.

Ref. [24] used a molecular technique (real-time PCR) to differentiate coffee blends, and arabica coffee presented amplification whilst robusta coffee did not. Thus, the detection of coffee species by means of real-time PCR is a promising technique for the further analysis of green and roasted coffee.

The above-reviewed works showed that a higher amount of Robusta coffee in blends leads to a lower acceptance of the drink. Due to the lower price of Robusta coffee (lower product costs and higher resistance) and the lack of information provided by some producers, fraudulent coffee is often encountered at the market. Blends of Arabica and Robusta coffee does not represent food fraud by themselves, but it is common to find premium or gourmet coffee with a higher composition of Robusta coffee than permitted by the regulations.

According to [24], in Brazil, the “Regulation on Minimum Standards of Quality for Roasted Coffee Beans and Roasted Ground Coffee” defines the composition of superior (premium) coffee, which may or may not contain Robusta coffee, limited to 15% of the total volume. On the other hand, another type of superior coffee is gourmet coffees, which are 100% Arabica coffee, from a single origin [28,29].

Thus, it is important to verify the amount of each species in the blend, to aid the food industry and consumers in avoiding fraudulent coffee. Research has been conducted with the goal of detecting the amount of Robusta coffee within blends, using techniques for detecting chemical components. Ref. [30] successfully used the content of P, Mn and Cu to discriminate Arabica and Robusta roasted coffee varieties.

Ref. [31] researched a model to predict the amount of Robusta and Arabica coffee within blends. They indicated that linoleic and α -linolenic acid were more abundant in Arabica coffee, while Robusta coffee contained a greater amount of oleic acid.

Ref. [32] verified that volatile organic compound (VOC) spectra were able to differentiate Arabica coffee from Robusta coffee, within green beans, roasted beans, ground coffee and brews. The authors concluded that VOC may be used throughout coffee processing, especially for roasted beans. Particularly for volatile compounds, Ref. [33] used a steam treatment as an alternative to improve the volatile profile and cup quality of coffee. According to these authors, the steam treatment increased the contents of acetoin, benzyl alcohol, maltol, 2,6-dimethylpyrazine, 2-furfurylthiol, and 5-methylfurfural, and decreased the contents of 4-ethylguaiacol, isovaleric acid, methional, 2,3-diethyl-5-methylpyrazine, and 3-methoxy-3-methylpyrazine. They indicated that a blend of 30% steamed coffee and 70% Arabica coffee was well accepted.

Ref. [34] applied infrared spectroscopy with photoacoustic detection (FTIR-PAS) to several blends between Arabica and Robusta coffee. The application of FTIR-PAS to coffee was able to characterize and classify blends, with the advantage of a sustainable, accurate, easy and quick method.

Differing from most of the research presented, [35] formulated a new type of blend: a mixture of 94% roasted coffee powder (Robusta and Arabica, 70/30, *w/w*), 3% cocoa powder, 2% coffee silverskin and 1% golden coffee (i.e., minimally processed green coffee). The authors concluded that this new blend had higher content of bioactive compounds and peculiar characteristics when compared with other commercial blends (Arabica and Robusta coffee).

From a sustainability point of view, coffee silverskin represents an environmental problem, produced mainly during coffee roasting. Nowadays, it is used as a fuel, as well as in composting and soil fertilization [36,37]. Due to its nutritional composition, research has been performed on the use of silverskin within coffee blends, as stated previously. Dietary fiber (56–62%), protein (19%), minerals (8% ash) and fat (1.6–3.3%) [36–38] are some of the chemical compounds of silverskin. In addition, phenolic compounds, such as chlorogenic acids (CGA) (1–6%), caffeine (0.8–1.25%) and melanoidins (17–23%) (Maillard reaction products), are also found in silverskin [39]. Depending upon the origin of the coffee and thus its silverskin, the chemical composition differs, as seen in Table 3.

Table 3. Nutritional composition of silverskin from six different geographical origins.

Geographical Origin	Nutritional Composition							
	Moisture (%)	Ash (%)	Fat (%)	Protein (%)	Insoluble Fiber (%)	Soluble Fiber (%)	Available Carbohydrate (%)	Caffeine (mg 100 g ⁻¹)
Cameroon	9.91	8.31	1.81	20.6	49.5	5.95	3.95	1154
India	10.30	7.34	1.19	18.9	50.6	9.00	2.70	676
Indonesia	9.28	8.71	2.46	18.2	47.5	7.55	6.35	1100
Brazil	9.53	10.4	3.15	16.7	44.2	11.20	4.80	1215
Vietnam	9.55	9.29	2.27	20.3	47.4	10.95	0.25	1140
Uganda	9.35	10.5	1.86	19.5	45.0	7.85	5.85	709

Source: Adapted from [39].

In Table 3, it can be seen why work is beginning to investigate mixing silverskin into coffee blends. Other procedures have also investigated to increase and/or mask the bitterness of Robusta coffee. Torrefacto coffee is one of them, which is produced by roasting whole beans with sucrose or glucose [40]. The addition of sugar forms a thin film on the beans' surface which protects the beans from oxidation and speeds up the Maillard reaction [40]. Thus, this addition is not intended to increase the sweetness of the coffee brew [26].

Ref. [10] investigated the influence of different coffee varieties and blends over the antioxidant activity. Coffee blends with high percentages of torrefacto roasts had stronger antioxidant activities.

Thus, in addition to blend composition and new product mixtures, the roasting and grinding processes have the potential to provide unique coffee flavors and aromas, according to the desired market. The development and innovative aspects of coffee production require the understanding of the blending ratio, which depends on the market. In other words, the sustainability of coffee production requires an understanding of species availability, the roasting degree, and consumer preferences.

3.2. Roasting and Grinding

The characteristic flavor and aroma of coffee result from a combination of hundreds of chemical compounds produced by the reactions that occur during roasting [41]. It is well known that roasting can be explained by three steps: drying, pyrolysis (roasting) and cooling. The first step removes water and volatile substances from the beans, and the color changes from green to yellow. The second step continues to remove water and volatile substances, along with CO₂, and the color changes to brown. This step is where several chemical reactions take place, including the Maillard and Strecker reactions, and the degradation of proteins, polysaccharides, trigonelline and chlorogenic acids [42]. Furans, pyrazines, pyrroles, and pyridines, among other substances that affect both the flavor and

aroma of the beverage, are formed from sugars and trigonelline [41]. Finally, cooling is required to prevent further oxidation (burning) of the beans. Figure 1 shows the color variation of coffee throughout the roasting process.



Figure 1. Coffee color throughout the roasting process [43].

Encouraged by the importance of the roast degree with regard to the sensorial characteristics of coffee, the Specialty Coffee Association (SCA), formerly the Specialty Coffee Association of America (SCAA), proposed a classification system of roasted beans by color, the SCA-Agtron [44]. In this classification, five color degrees of the beans are present, while allowing intermediate classifications between very dark, dark, medium, light and very light.

The medium light and moderately dark roast degrees are the most commonly used commercially, affecting the flavor and aroma of beans [45]. Depending upon the roasting degree, different cup qualities may occur, due to the development of different chemical com-

ponents during roasting (Table 4). Thus, different studies have assessed the acceptability and/or assessment of the chemical composition of roasted coffee.

Table 4. Characteristics of coffee beans/brews according to the roasting degree.

Roasting	Weight Loss (%)	Agtron Number	Bean Temperature (°C)	Characteristics
Cinnamon	13.0	80–75	90–130	Volatile compounds start to expand the beans.
American	14.0	74–65	170–190	First crack. Acidity higher than sugar.
City	15.0	64–60	210–220	First crack ends.
Full City	16.5	60–50	224–230	Second crack. Balance between acidity and sugar. Oils start to appear.
Vienna	17.0	49–45	230–235	Second crack ends. Lower acidity.
Espresso	18.0	44–35	235–240	Black with oil stains. Shiny surface. Sweet bitterness overpowers acidity.
French	19.0	34–25	240–246	Caramelization of sugars. Decrease in acidity. Burning smell
Italian	20.0	24–15	246–265	Loss of flavor. Shiny surface (oil).

Source: Adapted from [46].

Ref. [41] studied the composition of green and roasted Arabica coffee with different cup qualities, namely soft, hard, rioysh and rio. The soft sample, of higher quality, presented higher protein levels, caffeine and lipid contents, before and after roasting. Acidity increased and pH levels decreased as the cup quality decreased.

Ref. [47] evaluated the acceptance of coffees of the types soft, hard and rio with different types of roasts (light, express and dark), with the aid of 65 consumers of coffee. The samples with a dark roast, independent of the coffee type, were largely preferred by consumers in relation to the color, aroma, and flavor attributes and overall.

Ref. [48] investigated the impact of the degree of roasting, grinding, and brewing on the evolution of coffee aroma in green coffee beans. The light roast was sweeter in all stages, and the darker roasts attained higher intensity of the typical ‘coffee’ attributes (coffee, roasted, burnt/acrid, and ashy/sooty) [47].

Ref. [49] determined polyphenolic compound and caffeine contents of Arabica and Robusta coffees with three roasting degrees: light, medium and dark. The highest contents of polyphenolic compounds and caffeine were achieved in coffees roasted in light roasting conditions, decreasing with intensified roasting.

Ref. [50] analyzed the free radical contents of coffee beans. Free radicals are precursors of colored products in roasted food. The authors stated that increasing the roasting time (roasting degree) led to an increment in free radical content. During storage, the free radical content increased, although this increase was lower in whole beans than in half and fully ground beans, for which the rate was similar [50].

Ref. [51] indicated that roasting resulted in the degradation of chlorogenic acid and the formation of melanoidins, and did not affect antioxidant activity. Blends that possess a higher percentage of Robusta coffee displayed higher caffeine contents, with greater antioxidant activity. The caffeine content, and its relationship with grinding extent, was studied by [52]. Greater grinding extents led to significantly higher caffeine contents.

Grinding devices also impact coffee composition, as stated by [53]. The elements Ba, Ca, Co, Fe and P can be significantly altered due to the type of milling process (ball, cryogenic and knife mills). The composition of the materials from which the mill devices are made also impacts the final coffee composition, and hence even the same type of mill can result in different kinds of contamination depending on the material used (e.g., steel, titanium, tungsten carbide) and the hardness and composition of the samples [53].

After roasting and grinding, coffee is subjected to brewing. According to [54], several variables can modify the in-cup coffee quality, including the contact time between the water and the ground coffee, the extraction time, the ground coffee/water ratio, the water temperature and pressure (for espresso coffee), the type of filter, and the boiling process.

Some investigations have been conducted regarding the grinding level and the brewing method. Ref. [54] studied the extraction method (espresso coffee, specialty espresso, caffè Firenze, Moka, V60, Cold Brew, Aeropress and French press) and the grinding level (fine, coarse). They used the same raw material; however, due to the extraction method and grinding level, different-quality cups of coffee were attained. Ref. [55] presented a review of some parameters regarding the physicochemical characteristics and flavor of coffee brews, such as the particle size (grinding degree) and extraction method.

4. Conclusions

The sustainability of the coffee production chain is influenced by the roasting degree, alongside grinding, as well the coffee species and varieties used for blending, directly impacting the nutritional and sensorial characteristics of the coffee drink. There are several works that indicate, in a singular manner, the effect of the roasting level, grinding degree and extraction method on the final product's quality. Works which correlate the roasting level, grinding degree and types of extraction method and their impact on the nutritional value and sensorial acceptance of coffee are absent or scarce. In addition, the production of several residues formed during coffee processing is a problem that science can and must work on to aid the industry and producers to properly treat these residues. Thus, future research needs to be performed regarding these trends.

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