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Coffee By-products Derived Resources. A Review

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ABSTRACT

Coffee is the most common beverage and there are two main different methods to process coffee cherries. Coffee husk is the main by-product of coffee processing by dry method and is available in large quantities throughout the years, but its main application has been limited to animal feeding or energy production. Most of the coffee husk is disposed in landfills or arable land, usually with no care of its fate and changes to the source of pollution, especially in developing countries. ~~Food industry by-product is one of the challenges~~Coffee husk can have several re-uses, ~~and~~ but it is important to have environment-friendly methods to change ~~the coffee husk~~it into usable material or material to be recycled in nature because of its ~~relatively high~~important content of organic matter, chemical nutrients, and secondary compounds. The aim of this review is to recollect the amounts and uses of the coffee industry by-products, giving emphasis to ~~the~~ its transformation into compost because of their large content of nutrients and the need to introduce high valuable organics into the soil.

***Key Words*:** Coffee-Husk, Component, Detoxification, Application, Waste-Management.

1. INTRODUCTION

Demand for coffee in the last 150 years was more than in the past not only because of the increased population and urban development but also because coffee has become one of the most consumed beverages in the world [1]. Nowadays coffee trade is economically at the second place of the world rank after petroleum [2]. According to the International Coffee Organization (ICO), in the 2016/2017 crop year the world coffee production was ≈152 million of 60-kilograms bags (for a total of ≈9 million tons), for an economic value of ≈90 billion dollars [3]. The world population of ≈7.6 billion in 2017 is expected to reach 8.6 billion in 2030, and 9.8 billion in 2050, with one third of the population concentrate in urban areas [4]. Because of this, also production and consumption of coffee are expected to increase concerning the actual levels.

Even though the first plantation of coffee was done in Yemen by Arab people in 13th century with seeds transferred from Ethiopia [5], nowadays Brazil, Vietnam, Indonesia, Colombia, Ethiopia, India, and Mexico are the major producers of coffee, with Brazil producing half of the world production [6]. The coffee plant belongs to the *Rubiaceae* family and, among the numerous species present in nature, currently only *Coffea arabica* L. (known as Arabica) and *Coffea canephora* L. (known as Robusta) have an important economic value [3]. Coffee processing industry produces huge amount of by-products since from 30 to 50% of coffee fruit weight is waste [6]. Due to the high amount of coffee seeds production, several re-using solutions have been proposed, but a win-win solution to manage the considerable amount of coffee husk is needed [7].

Because of this, the aims of this review were to report of 1) different processing methods of coffee cherries, introduce the main by-products of coffee processing, and emphasize on the needed detoxification of coffee husk obtained by dry method; and 2) the several usages of coffee husk in industry and agriculture. All the themes referring to processes and uses were discussed also by synthesizing advantages and disadvantages reported in the literature.

2. COFFEE SEED ANATOMY

The economic lifespan of coffee tree is maximum of 30 years. The shrub is perennial and can reach a height of 10 meters. The first flowers are produced when plants are 3-4 years old, and are creamy white and sweetly scented, appearing in clusters in the axis of the leaves. Two fertilized ovules of coffee flower ovary start to grow up two months after fertilization. Adequate water supply is important to break the dormancy in the third month. The ovary size increases, and the embryotic sac grows and fills with endosperm. Till the end of the fifth month after fertilization, weight and volume of fruits increase significantly. Between sixth and eighth months after fertilization, the fruit reach to maturity, represented by an oval drupe of 18 mm in length and 10-15 mm in diameter. The ripe fruit has bright red or yellow color, and it is also called “cherry” [8][9][10].

The coffee fruit has four different layers protecting the seed that must be removed to collect the two beans forming the seed. The outer layer is the skin (epicarp or exocarp), with waxy substance and red color. The second layer is the pulp (mesocarp), that is a slim layer of pectinaceous materials. The third layer is the parchment (endocarp), with polysaccharide covering. The last layer, sticking to the seed, is named silverskin or chaff [11].

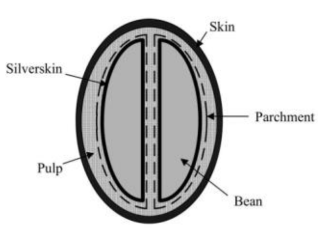

Figure1: Schematic picture of coffee fruit from [11].

3. COFFEE PROCESSING METHODS AND BY-PRODUCTS

To keep the quality of the seeds and preserve them from pathogens, they must be extracted from the four layers forming the other part of the fruit. The industrial process of coffee seeds preparation can be made following two main methods: dry and wet methods. The dry method is the traditional one, but it is also the simplest and environment-friendly since it produces less amount of solid and liquid by-product. Following this method, after having selected and cleaned the cherries, these are sun-dried with frequent turning to obtain a relatively homogenous drying. Thus, the outer layers of the cherries are removed by a hulling machine, and the coffee beans are roasted and bagged. With the wet method (or washed method) more equipment and water are needed in comparison to the dry method. With this method, the quality of the coffee beans is higher than with the dry method because the bean components are better preserved, and the number of defective seeds is less. Following this method, after the cherries are sorted and cleaned, the pulp is separated by a squeezing machine and the seeds are roasted [3]. Every ton of fresh coffee cherry produces 0.12-0.18 tons of coffee husk with the dry method and 0.5 tons of coffee pulp with the wet method [12]. Even though the quality of the obtained coffee seeds depends on the processing, the seeds produced with both methods have their own market since the beverage obtained with seeds submitted to the dry process is less acidic than those obtained with the wet method.

Each step of the coffee processing from coffee fruit to a cup of coffee, including separation of coffee seeds, roasting, packing, and making a drink, produces by-products.

3.1. Spent coffee ground. This by-product is the result of coffee brewing from coffee making such as homemade coffee and coffee machines or indirect way like instant coffee and beverage factories. It has a dark brown color, coarse texture, and high moisture [11]. The content of lipids in the fresh spent coffee ground is around 2% on a weight basis, with palmitic and linoleic acids covering 35% of the total extractable oil [13]. This by-product is also rich of vitamin E since, in classic espresso coffee and in coffee machines coffee, only 1 and 5%, respectively, of this vitamin is extracted. Therefore, coffee ground cakes can be used as a source of liposoluble antioxidant vitamins [14].

3.2. Defective and premature coffee beans. Both coffee harvesting and roasting process produce two types of by-products, respectively: immature and defective beans. These beans must be removed from the mass of the valuable beans since they might decrease the quality of the final products [15]. In fact, beans from defective cherries have higher amounts of free amino-acids and phenols and contain fewer sugars than normal beans because they did not reach proper maturity [16]. As an alternative use for these low-grade coffee beans, Alves *et al.* [11] suggested to use them for the extraction of chlorogenic acid or caffeine, for their potential applications in the food and pharmaceutical sectors.

3.3. Silverskin. Coffee silverskin is a so thin layer sticking to the coffee seeds that detaches only during coffee roasting [17]. Coffee silverskin has antioxidant activity, because of the presence of melanoidins [18], prebiotic activity [19], and contains dietary fiber [20]. These valuable components have encouraged studies on the production of body weight control beverages, diet bread, biscuits [17][21], and cosmetic products [22].

3.4. Coffee pulp. Coffee pulp represents ≈35% of the coffee fruit [11] and is a by-product of the wet method in coffee hulling process. In the coffee pulp, the content of phenolic acids is slightly higher than in coffee husk, 1.5% *vs*. 1.2%. Among the phenolic acids comprising the coffee pulp, flavan-3-ols, hydroxycinnamic acids, flavonols, and anthocyanidins are the most represented [23].

3.5. Coffee husk. Coffee husk is the main by-product of the dry method and is formed by all the layers at once, including dried skin, pulp, mucilage, and the parchment [24]. When the coffee cherry is dried, ≈12-18% of the dried fruit weight is coffee husk [5]. In general, amount of components and indexes of coffee husk vary according to the coffee species, the geographical origin of the cherries, and the chosen method of processing [25], which explains the differences in the composition reported by many authors [26][2][27][11][6][15]. ~~For example, in~~In Table 1 we reviewed ~~, compared~~ and synthesized composition and physicochemical properties of coffee husk. However, ~~taking into consideration the values reported in Table 1,~~ Alves *et al.* [11] reported completely different amounts of lignocellulosic polymers, with 24.5% cellulose, 29.7% hemicellulose, and 23.7% lignin. It is desirable that in future studies coffee husks be classified according to their properties.

Table 1: Main composition and physicochemical properties of coffee husk.

	Value	References
Organic component (g kg^{-1})		
Carbohydrates	580-850	[27][6][11]
Cellulose	430	[2][27]

Hemicellulose	70	[2][27]
Lipids	5-30	[27][6][11]
Total fiber	240	[2]
Ash	25-62	[28][11]
Protein	80-110	[27][6][11]
Caffeine	10	[2][6][11]
Tannins	50	[2][6][11]
Chlorogenic acid	25	[2]
Pectic substance	16	[2]
Lignins	90	[2]
Sugar content (g kg^{-1})		
Reducing sugar	120	[29]
Total sugar	140	[29]
Sucrose	20	[29]
Physicochemical parameters		
pH (1:10)	5.35-6.63	[30][31]
EC (dS m^{-1})	2.24-3.1	[30][31]
Organic carbon (g kg^{-1})	545	[30]
Organic matter (g kg^{-1})	815	[31]
C/N ratio	29.8-40	[30][31]

3.5.1 Macro- and micro-nutrients. Coffee husk is rich of macro and micro-nutrients, with considerable amount of N (1720-1830 mg kg^{-1}), P (80 mg kg^{-1}), K (20 600 mg kg^{-1}), and others (Table 2). Positively enough, it contains small amounts of Na.

Table 2: Elemental content of coffee husk. Values are expressed on a dry matter basis.

Element (mg kg^{-1})	Coffee Husk	References
Total content of inorganic elements	5000-30 000	[32]
N	1720-1830	[32][30]
P	80	[32]
K	20 600	[32]
Ca	2210	[32]
Mg	790	[32]

Fe	260	[32]
Cu	20	[32]
Mn	60	[32]
Zn	10	[32]
B	91.4	[33]
S	1100	[33]
Se	0.19	[32]
Na	40	[32]

3.5.2 Amino acids. The coffee husk contains a protein content ranging from 8 to 11 % on a dry matter basis [34], with a relatively high content of amino acids such as glutamic acid (7.7% of the total protein content) and aspartic acid (7.1%) [34]. Glutamic acid is responsible for the transport of glutamine and other amino acids through the blood, and its presence decreases the need to consume sugar and alcoholic beverages. The aspartic acid is involved in the metabolism of DNA and RNA, but also in protecting the liver and boosting the immune system. So, coffee industry by-products are a source of amino acids that could be evaluated as dietary phytochemicals useful for human beings. Dietary supplements and/or food fortification based on coffee by-product production may be feasible too [35][36]. Table 3 shows a comprehensive view of the main amino acids present in the coffee husk.

Table 3: Content of protein and of the main amino acids in coffee husk and pulp. From [27] [34].

Protein content	8-11%, on a dry matter content
Amino acid	% with respect to the total protein content
Glutamic acid	7.7
Aspartic acid	7.1
Leucine	4.7
Glycine	4.2
Proline	3.7
Valine	3.7

Alanine	3.5
Lysine	3.4
Serine	3.3
Isoleucine	3.3
Threonine	3.1
Phenylalanine	3.0
Arginine	2.8
Histidine	2.5
Tyrosine	1.9
Methionine	0.3
Cystine	0.3

3.5.3 Volatile oils. Al-Yousef and Amina [37], working on coffee husk from *Coffea arabica* L., reported of the content of a volatile oil made of at least 55 molecules. As reported in Table 4, the main chemical compositions of volatile oil in the essential oil of coffee husk, as determined by gas chromatography-mass spectroscopy (GC-MS), is mainly represented by butylatedhydroxy (65.83%), with much smaller content of 1,2-benzenedicarboxilic acid (7.28%), phenylethyl alcohol and octanoic acid (1.69% each), and 2,3-isopropylidene-6-decoxyhexo (1.63%). According to the mass spectra observation, 30% and 40% of the compounds present in the oil are hydrocarbon and oxygenated constituents, respectively, while aromatic compounds dominates. Volatile components showed antibacterial, antifungal, and antioxidant potentiality that are helpful in the treatment of infection diseases. In Table 4, the total time required to analyse a single sample was 58 min~~,~~ and the components were identified on the basic of GC-MS retention time. M+ represents molecular ions, which are important for determining the molecular weight by GC-MS.

Table 4: Content of the main volatile oils in the essential oil of coffee husks. From [37].

	Required time (min)	Area (%)	M^+ (g)

Butylated hydroxytoluene	24.2	65.83	220.18
1,2-benzenedicarboxylic acid	31.92	7.28	278.34
Phenylethyl alcohol	13.66	1.69	122.09
Octanoic acid	15.76	1.69	144.21
2,3-isopropylidene-6-deoxyhexo	26.12	1.63	220
Decane, 1,1'-oxybis-	47.9	1.59	298.54
Nonanoic acid	18.4	1.58	158.16
1,2-benzenedicarboxylic acid	33.78	1.37	278.35
Beta-d-arabino-2-hexulopyran	24.66	1.17	234.00
Oxalic acid, 2-ethylhexyl tetr	44.78	1.11	398.61
~~h~~Hexatriacontane	49.36	1.00	506.97

M+ is the molecular ion, expressed as the ratio between mas and charge number of ions (M/Z); since Z is almost always 1 in GC-MS, M+ is mainly generally the mass (g) of the ionic molecule

Al-Yousef and Amina [37], evaluated the volatile oil and total alcohol extract of coffee husk for their antimicrobial activity with respect to three well-known antibiotics like ~~. A~~ampicillin and ~~Doxycycline~~ doxycycline, ~~were~~ used as positive control against bacteria, ~~while~~ and ~~Nystatin~~ nystatin, ~~was~~ used as the control antifungal drug. In the experiment, minimum inhibitory concentration of ethanol extracts as well as volatile oil of coffee husk against drug resistant clinical strains was determined. The results are shown in Table 5. Both volatile oil and total alcohol extract of coffee husk reduced the growth of *Staphylococcus aureus*, *Escherichia coli*, *Pseudomonas aeruginosa*, and *Candida albicans* colonies by 50 to 104% with respect to three tested antibiotics, with the alcohol extract being more efficient than volatile oil for *Staphylococcus aureus* and *Pseudomonas aeruginosa*.

Table 5. Antimicrobial activity and minimum inhibitory concentration of essential oil and total alcohol extract of coffee husk from *Coffea arabica* L. on the growth of four infective microbes [37].

Samples	*Staphylococcus aureus*	*Escherichia coli*	*Pseudomonas aeruginosa*	*Candida albicans*
	Zone of inhibition (mm)			
Coffee husk essential oil	14.0 ± 1.3 (-66.7%)	17.0 ± 1.9 (-68.0%)	13.0 ± 2.0 (-50.0%)	15.0 ± 1.3 (-5.2%)

Total alcohol extract	22.0 ± 0.5 (-104.8%)	17.0 ± 2.5 (-68.0%)	20.0 ± 1.1 (-83.3%)	14.0 ± 1.2 (-0.9%)
Ampicillin	21	-	-	-
Doxycycline	-	25	24	-
Nystatin	-	-	-	23
	Minimum inhibitory concentration (mg mL^{-1})			
Samples	*Staphylococcus aureus*	*Escherichia coli*	*Pseudomonas aeruginosa*	*Candida albicans*
Coffee husk essential oil	0.8	0.8	0.8	0.8
Total alcohol of coffee husk	3.2	>3.2	3.2	>3.2

*Values of Minimum inhibitory concentration is given in % v/v for dry oils

3.5.4 Phenolic acids. The main phenolic acids of fresh coffee husk are caffeine, tannins, and chlorogenic acids (for quantities see Table 1), whose presence prevents various uses of fresh coffee husk because of their ecotoxicological concerns [38]. For example, fresh coffee husk is not suitable for animal feeding because of its anti-nutritional properties due to the excess of phenolic acids [39]. Thus, chlorogenic acid has phytotoxic effects able to decrease seed germination and plant growth and, because of this, it cannot be distributed in soil as soil fertilizer [40][38]. In addition, caffeine and tannins negatively affect aquatic organisms like algae, sea urchin, and fishes, which develop morphological and behavioral abnormalities [41].

Caffeine. The alkaloid caffeine (1,3,7-trimethylxanthine) has been found in more than 60 plant species, with the highest levels in coffee beans, tea, and cocoa. Other two alkaloids of the xanthine derivative group are theobromine (3,7-dimethylxanthine) and theophylline (1,3-dimethylxanthine) [42]. Generally, caffeine has positive effects on humans as it has chemical structure like that of adenosine, so it is well-known as adenosine receptor. Because of this, caffeine may help to be relaxed and sleep [43]. Caffeine can be also able to contrast obesity and diabetes [44], as well as Parkinson's [45] and Alzheimer's symptoms [46]. However, caffeine has negative effects on the environment as it is toxic to aquatic organisms and mammalians, and has negative effects on animals, plants, fungal and bacterial growth [47].

Tannins. Tannins are commonly found in the bark of vascular plants and, to a lesser extent, into leaves, fruit, flowers, and seeds [48]. Tannins are considered as anti-nutritional compound, and this aspect limits the use of coffee husk in animal feed [49]. Benefits of tannins for human health include antibacterial and antifungal activity, ~~which destroys and prevents the development of bacteria and fungi~~ [50], antimicrobial activity, ~~which is~~being effective against ~~bacteria, fungi,~~ parasites~~,~~ and some viruses [51], ~~and~~ anti-inflammatory [52], and anti-allergy [53] activities. Tannins are also known for their low biodegradability; because of this reason they tend to remain for long time in the environment and accumulates in the food chain [54].

Chlorogenic acid. The esterification of caffeic acid with quinic acid produces chlorogenic acid, which is a soluble polyphenol [55] that plays many human health benefits, including neuronal cell death protection [56] and anticancer activity [57][58]. Chlorogenic acid plays positive roles also in plant functions including cell wall synthesis, wound healing, and root hair formation [59]. However, depending on its concentration, it may play a negative role especially in roots [60]. Villarino *et al.* [61], reported the inhibitory effect of chlorogenic acid on fungal growth, due to its role on plant defence. The content of other phenolic acids extracted from coffee husk is reported in Table 6.

Table 6. Phenolic components of coffee husk (modified from [62]).

Method	Condition/Solvent	Epicatechin	Gallic acid	Tannic acid	Protocatechuic acid	Vanillic acid
		μg of gallic acid equivalent per kg of coffee husk (dry matter)				
Ultrasonication	Ethanol	-	-	-	-	2346.7
Soxhlet	Ethyl acetate	47.6	3869.2	-	-	-
	Ethanol	-	-	3859.2	-	-
SFE*	200bar/40^{0}C	-	14.85	-	-	-
CO_2	300bar/40^{0}C	32.55	-	-	12.4	-

* SFE: supercritical fluid extraction.

Secondary metabolites in coffee husk such as caffeine and other phenolic compounds are good source of antioxidants. Table 7 shows the antioxidant capacity of aqueous extract of coffee husk evaluated following DPPH (2,2-diphenyl-1-picryl-hydrazyl-hydrate) radical sequestration method and inhibition of co-oxidation of β-carotene and linoleic acid percentage [63]. For this experiment, coffee fruits were randomly collected at four different farm locations, from two plants in the northern (husk 1 and grains 1), southern (husk 2 and grains 2), eastern (husk 3 and grains 3), western (husk 4 and grains 4) and central region (husk 5 and grains 5) of the plantation [63]. Results indicated that micro-environmental conditions present in the plantation affect the antioxidant capacity of aqueous extract of coffee husk.

Table 7. Antioxidant capacity of aqueous extracts of coffee husk evaluated by the free DPPH radical sequestration method and inhibition of co-oxidation of β-carotene and linoleic acid [63].

Sample	DPPH EC50* (mg mL^{-1})	Inhibition of co-oxidation of β-carotene and linoleic acid (%)
Coffee Husk 1	4.71^{f}	40.78^{bc}
Coffee Husk 2	3.57^{h}	34.88^{c}
Coffee Husk 3	4.44^{g}	43.74^{abc}
Coffee Husk 4	2.73^{j}	44.55^{abc}
Coffee Husk 5	3.44^{i}	40.80^{bc}
Coffee Grain 1	15.09^{a}	68.58^{a}
Coffee Grain 2	11.48^{b}	66.43^{ab}
Coffee Grain 3	10.44^{c}	58.22^{abc}
Coffee Grain 4	10.10^{d}	64.65^{ab}
Coffee Grain 5	7.53^{e}	68.22^{a}

* EC50 = half maximal effective concentration.
For each column, means followed by different letters differed for $P < 0.05$, by the Tukey test.

3.5.5 Lignocellulosic materials. Cellulose, hemicellulose and lignin are the principal lignocellulosic components forming plant cell walls. Lignocellulosic compounds like phenolic acids often prevent coffee husk usage and degradations, so it is necessary to find techniques able to break down these substances ~~prevention and let these materials to recycle quickly~~ [64][65]. According to Oliveira *et al.* [66], the lignin of coffee husk represents a significant resource for enabling use of coffee husk as raw material for ~~emerging~~ biorefineries where lignin can be separated from other coffee husk components with a pre-treatment ~~with~~ by diluted acid followed by soda extraction. ~~Also, the~~ The extracted lignin can be then wet-oxidized under aqueous and alkaline conditions, in order to produce valuable products such as low molecular weight biochemicals.

Many factors, like lignin content, crystallinity of cellulose, and particle size, limit the digestibility of hemicellulose and cellulose. Pre-treatments improve the digestibility of the lignocellulosic material. Each pre-treatment has its own effect(s) on ~~the~~ cellulose, hemicellulose, and lignin. Many thermal, acid, alkaline, and oxidative pre-treatments~~-~~ have been evaluated for improving biodegradability of lignocellulose substrates [64]. For example, Baêta *et al.* [67] pre-treated coffee husk by a steam explosion technique that increased the bioavailability and biodegradability of cellulose,~~–~~ broke down the lignocellulose structural components, and produced soluble organic compounds. This method is effective for increasing the anaerobic biodegradability too.

3.5.6 Flavorings. The most important characters of coffee as a beverage are acidity, aroma, and taste. Without acidity, the coffee is approximately tasteless [68]. Sampaio *et al.* [69] reported that coffee husk is a valuable by-product due to its aroma and presence of sugars that can be converted to ethanol. Table 8 shows flavor and aroma inside beverages made by different concentration of coffee husk and pineapple juice.

Table 8. The sensorial analysis of flavor, aroma and overall appearance of the beverage developed with the rinds of coffee with different concentrations and pineapple juice [63].

Sample	Flavor	Aroma	Overall impression
	Score		
F1*	2.96 ± 1.88[b]	4.25 ± 2.06[b]	3.75 ± 1.90[b]
F2**	4.86 ± 2.02[a]	5.44 ± 1.91[a]	5.05 ± 2.01[a]
F3***	5.48 ± 2.11[a]	5.44 ± 1.95[a]	5.48 ± 2.05[a]

For each column, means followed by different letters differ for *P* < 0.05, by the Tukey test.
*100% coffee husk extract; **90% coffee husk extract+10% concentrated pineapple juice; ***80% coffee husk extract+20% concentrated pineapple juice. The evaluation was assessed by non-trained 52 judges who used a structured 9-point hedonic scale (1 = I greatly dislike, 9 = I enjoyed it very much).

According to Tables 7 and 8, aqueous extracts of coffee husk represent a promising natural source of bioactive phytochemicals, also because of their low levels of antinutrients [63]. Neves *et al.* [63] noticed that the beverage incorporated with concentrated pineapple juice presented the greatest acceptability, besides increasing the antioxidant capacity of the product. Thus, the formulated beverages constitute a promising alternative for the beverage market, given the meaningful content of phenolic constituents derived from coffee husk.

3.5.7 Detoxification. Phytotoxic compounds like caffeine, chlorogenic acid, and tannins (Table 1), if released into the environment from coffee waste, can have severe ecotoxicological ~~effects. Although there are many benefits of caffeine consumption to human health, studies indicate that environmental leaching of caffeine has~~ ~~detrimental~~ effects on several organisms [70]. Therefore, detoxification of coffee husk~~s~~ from phytotoxic compounds and antinutritional factors, or at least degrading them to a plausibly safe level for reusing or recycling, is necessary. Detoxification of coffee husk with physical, chemical, or biological methods were studied by [71][72][42], while a general review of enzymatic and microbial methods to remove caffeine is reported by [73].

Some physical (percolation), chemical (alcohol extraction), or microbial (fermentation with fungi) treatment~~(s)~~ can reduce the phenolic content in coffee husk. ~~Among all the systems to reduce the toxic effect of coffee husk, t~~Treatments with bacteria and/or fungi~~,~~ and composting are the most used treatments for coffee husk~~,~~ and for other coffee by-products like coffee pulp and silverskin because they are more efficient and economic~~al~~ for controlling huge amount of waste. High concentrations of bacteria are required for caffeine detoxification since caffeine has a toxic effect for bacteria, and 0.1% concentration of caffeine inhibits protein synthesis in bacteria and yeast [71][74]. However, some microorganisms can grow in presence of caffeine and survival is due to their capacity to degrade it [75][76]. Several studies were carried out to investigate the use of purines, including caffeine, as a source of energy for microorganism growth [77]. Although fungi growing on caffeine have been isolated, most of the studies were done with bacteria isolated from soil, mainly belonging to the *Pseudomonas* group, with emphasis on *Pseudomonas putida* [78]. Yamaoka-Yano and Mazzafera [79] used *P. putida* strain and, after a short incubation periods of 9 days, observed 40% reduction of caffeine. Brand et al. [71] tested biological detoxification of coffee husk by filamentous fungi (*Rhizopus*, *Phanerochaete*, and *Aspergillus* spp.) using a solid-state fermentation system in which coffee husk was used as the sole source of C and N. *Rhizopus arrizus* LPB-79 strain showed great results on caffeine and tannins degradation (87% and 65%, respectively), which were obtained in 6 days at pH 6.0 and at 60% moisture.

The toxicity of coffee leachate were studied in laboratory by standardised toxicity tests on aquatic organisms [80][7], and results showed that the half maximal effective concentration (EC50) of coffee leachate was 6.02% v/v on the bacterium *Vibrio fischeri*, lower for the bacterium *Daphnia similis* (~~EC50 of~~ 1.5%), and even less for the microcrustacean *Ceriodaphnia dubia* (~~EC50 of~~ 0.12%). The reduced EC50 values from bacteria to water fleas was explain as the result of increased exposure to ingestion. There are good studies on caffeine toxicity, but no toxicity test has been performed on leachate from coffee by-products~~ers~~. Furthermore, there are

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ha formattato: Tipo di carattere: Corsivo several paths through which coffee can enter the environment such as processing/roasting or the retail consumption, suggesting that there is a major gap in toxicity data for coffee industry by-products that requires urgent attention [7].

4. USES OF COFFEE HUSK

Coffee husk is the main coffee by-product that has been the topic of several studies in order to use it in industrial activities, to produce biofuel, as contaminants sorbent, dietary fiber, and bioactive compounds, for the extraction of enzymes, or in agriculture as animal feeding or for making compost, silage, biochar, or mushroom substrata.

4.1. INDUSTRIAL USES

4.1.1. Coffee husk in ceramic industry. Generally, coffee producing countries use coffee husk as solid fuel and this method produces huge amount of ash~~es~~ that has environmentally side effects. Ash~~es~~ obtained by coffee husk combustion (that collected from ash dumps) are rich in alkaline and alkaline-earth metals that are a candidate ~~of~~ for replacing the scarce and expensive feldspars traditionally used as fluxing component in clay based ceramic formulation. Results shows that adding 25-40 ~~wt.~~% of ashes in common clay-based ceramic formulation had the best result in ceramic quality [81].

4.1.2. Coffee husk in particleboards production. Bekalo and Reinhardt [26] and Nuamsrinuan *et al.* [82] studied the use of coffee husk for partial replacement of wood (up to 50%) in the production of particleboards. ~~They tested for the mechanical properties, swelling and water absorption standards.~~ The results of particle sheet from milling process passed the standard tests of mechanical properties, ~~but~~ while swelling and water absorption did not~~indicated that all the~~

~~coffee husk sheet were not passed it~~. ~~The effect of coffee husk particle size and isocyanate adhesive on mechanical properties was clarified.~~ The coffee husk-wood board showed great promise for its use in structural and nonstructural panel products based on superior flexural and internal bond properties.

4.1.3. Flavor production. Food flavoring compounds can be produced by chemical synthesis or extracted from natural materials. Nowadays the second way is highly demanding since the obtained products are considered safer and healthier than those obtained via synthesis. Plants are acceptable source of essential oils and flavors, but their value ~~also~~ depends on factors like weather conditions and plant diseases. Due to the presence of anti-nutritional factors such as caffeine and tannins, coffee husk cannot be used directly as a flavour source [83]. Instead, when coffee husk is treated by steam to remove caffeine and chlorogenic acids, it can be used for aroma production by using fungi of the genus *Ceratocystis* [83]. Soares *et al.* [83] tested coffee husk as raw material for fruits flavor production by solid state fermentation and found that different dosages of glucose can determine the production of different flavors such as banana and pineapple.

4.2. FUEL PRODUCTION

4.2.1. Coffee husk as a solid fuel. The use of coffee husk as solid fuel is the simplest way to manage problems due to disposal or accumulation in nature, even though the production of ash is also raising concerns. In fact, the ash derived from coffee husk combustion is often the object of illegal covert disposal and the source of environmental impacts [81]. About 70% of the coffee husks produced in Kenya is used as solid fuel [84]. The coffee husk is carbonized in a kiln and then ground, coagulated, and molded in form of briquettes prior to being packed into bags. The obtained coffee charcoal briquettes have better quality than wood charcoal [29][68] but, as for other agricultural residues, carbonization is not the best choice to recover energy from coffee husk, as its combustion efficiency is minimal because of not exactly suitable physicochemical properties such as low bulk density, low ash melting point, and high volatile matter content [85].

4.2.2. Gasification of coffee husk. In order to find solutions to improve energy recovery from coffee husk, gasification is a possibility to increase energy recovering by producing ignitable gas through a partial incineration at elevated temperatures and moderate heating rates. The obtained gas is a mixture of CO, H_2, CH_4, CO_2, and N_2, and temperature level is key to improve gas quality [86]. In coffee producing countries, biomass energy has potential to be the most abundant sustainable renewable energy but, to reach this goal, there is ~~a~~ the necessity to develop and sustain contemporary technologies that increase the biomass-to-energy conversion. One way can be the high temperature air/steam gasification of biomass [87]. Wilson *et al.* [87] studied coffee husk experimental gasification under high temperature conditions by batch facility and found positive influence of high temperature on increasing the gasification process. Experiments carried out at 4% O_2 concentration obtained the highest gasification rate (96% of the coffee husk), while 82.80% and 71.29% of the husk was gasified with gasification conditions at 2 and 3%, respectively. Miito and Banadda [86] found that the 46.6 million tons per year of coffee husk produced in Uganda, with a heating value of 18.34 MJ kg^{-1}, will address a 0.7% of the total energy consumed in the country, while protecting the environment too. The same use could be feasible in the countries where coffee is produced and processed, namely where coffee husk abounds.

4.2.3. Ethanol production from coffee husk. Coffee husk has also good potential to be used for bio-ethanol production and, as for gasification, temperature and yeast concentration are key to control the quality of the production with batch fermentation method [88][27]. The availability of cellulose, hemicellulose, and lignin in coffee husk is similar to that of other agricultural residues such as sugarcane bagasse, barley and wheat straws, rice husk, and others. ~~But~~However, because of the high amount of coffee husk generated, the toxic nature of coffee husk, the high percentage of fermentable sugar, and the presence of high concentration of carbohydrates, it could be a good source of raw material for bio-alcohol production [89].

4.3. ADSORPTION OF CONTAMINANTS

Cu, Cr, Cd, Ni, Hg, Pb, and Zn are the most abundant pollutants in industrial wastewater. Common methods for the removal of heavy metals from wastewaters include ion-exchange, filtration, electrochemical treatment, chemical precipitation, and adsorption. Since the activated charcoal used to remove organic and inorganic pollutants from aqueous effluents is expensive, and the activated charcoal produced from coffee husk showed high specific surface and porosity [90], it is a valid solution to reduce costs for wastewater treatment [91][92]. In addition, the adsorbed metals can easily desorb and the biomass be ready for final disposal [93]. Adsorption of Pb [94], Ni [95], cyanide [96], dye contaminants [97], and antibiotic norfloxacin [98] by coffee husk in batch mode is used to decontaminate aqueous solution. Berhe *et al*. [93] studied the efficiency of coffee husk to adsorb Pb(II) from industrial effluents using batch experiment and found that, at optimum adsorption conditions ~~adsorption~~ (pH 5 and~~-~~ 90 min of contact time at 200 rpm), there was the maximum adsorption efficiency of 95.14%.

4.4. PRODUCTS OBTAINED BY FERMENTATION

4.4.1. Organic acids. Coffee husk is a cheap and available substrate to produce organic acids like gibberellin and citric acid by fermentation techniques. Shankaranand and Lonsane [99] produced citric acid from coffee husk by using *Aspergillus niger* under solid state fermentation method, and by every 10 g of coffee husk they produced 1.5 g of citric acids. Machado *et al.* [100] evaluated the feasibility of employing coffee husk as a substrate to produce gibberellic acid in both solid-state fermentation and submerged fermentation tests.

4.4.2. Enzymes. Coffee by-products can be also used to produce enzymes like pectinase, tannase, and caffeinase by two main industrial enzyme production methods: solid-state fermentation and submerged fermentation [101]. However, Battestin and Macedo [102] produced tannase from coffee husk by using *Paecilomyces variotti,* while Murthy and Naidu [101] studied the production of amylase, protease, and xylanase by fungal organisms.

4.5. BIOACTIVE COMPOUNDS

Bioactive compounds are an extra nutritional factor typically present in small quantities in foods that have been intensively studied to evaluate their effects on health. Some of those are phenolic compounds or antibiotic molecules, while other have an anti-inflammatory, hepatoprotective, and antioxidant activity, or the ability to improve cognitive capabilities [7]. Agro-industrial by-products are good sources of bioactive compounds and have been explored as sources of natural antioxidants [103].

4.5.1. Dietary fiber. Agro-wastes are great sources of dietary fiber, which include cellulose, hemicelluloses, lignin, pectin, gums, and other polysaccharides. The soluble and insoluble dietary fibers have a wide range of health benefits, such as reduction of the risks of gastrointestinal diseases, cardiovascular diseases, and obesity [104]. The kind of coffee and the degree of roasting and extraction method influence the dietary fiber content and structural characterization of coffee husk and other coffee by-products, as determined by the Association of Official Agricultural Chemistry ~~.~~ (AOAC) methods ~~for the determination of dietary fiber is well-known as a global standard method used for labelling food products~~ [105].

4.5.2. Anthocyanins. Anthocyanins are flavonoid compounds responsible for the red/blue color of many fruits and flowers. By using concentrated methanol as extractant, Prata and Oliveira [106] reported cyanidin 3-rutinoside as the dominant anthocyanin in coffee husk, so ~~it~~ this latter could be used as a source for anthocyanin pigments as natural food colorant. ~~Anthocyanins is extracted by concentrated methanolic from coffee husk.~~

4.6. AGRICULTURE

Uses of coffee husk in agriculture can be many, but the high content of phenolic acids and the mutagenic effect of caffeine suggest that recycling of coffee husk in agriculture should be preceded by detoxification process(es) able to decrease the concentration of these components. Information about detoxification of coffee husk is given at point 3.5.7.

4.6.1. Animal food. Agricultural industry by-products like coffee husk as livestock food is important to reduce the food competition and help to environment sustainability. Yearly coffee roasting industry produces million tons of coffee husks that contain valuable nutrients like proteins, carbohydrates, and minerals. Huge amount of production and good nutrients content make coffee husk a good material for animal food [107]. However, the idea of using coffee husk for ruminants, pigs, chickens, fishes, and rabbits was released several decades ago, but the result was not so bright and acceptable. In fact, National Dairy Board [108] reported that coffee husk is not a delicious food for cattle, which can tolerate only small portion of it because of the content of phenolic components. Fishes and poultries are even more sensitive than cattle and pigs, so the quantity of coffee husk in their diet must be small [34], unless to submit the husk to a detoxification process.

4.6.2. Mushroom bed. Coffee husk is an appropriate bed for mushroom growth because of its availability and cheap price and, due to its fragmented nature, no grinding is needed before application, but it needs disinfection. Coffee husk is a good substrate for mushroom bed, especially for *Lentinula edodes* (shiitake) and *Flammulina velutipes* species [109]. Fermentation of coffee husk by the fungus *Pleurotus ostreatus* increased protein and cellulose contents and decreased the proportion of lignin, tannins, and caffeine. Further, when fermentation of coffee husk increased, the volatile fatty acid and digestible dry matter decreased [109].

4.6.3. Biochar. Biochar is charcoal produced by pyrolysis of organic materials and can be used as soil amendment and fertilizer [110]. The biochar quality depends on the nature of the raw material and temperature. Acid soils, which abound in tropical areas, have deficiencies in plant nutrients like N, P, K, Ca, and Mg, and consequently have low crop production rates. Adding biochar reduces the soil acidity due to the alkalinity of biochar and increases the availability of nutrients and water [111]. Studies on coffee husk biochar showed that it improved soil chemical properties by increasing pH, electrical conductivity, cation exchange capacity, organic matter, total N , and available P [112]. Dume *et al.* [113] reported that the application of 15 tons ha^{-1} coffee husk biochar that had been produced at 500^0C temperature had positive result on soil fertility and yield. Deal *et al.* [114] compared the performance of biochar in five different feedstocks (coffee husk, maize cobs, eucalyptus wood, groundnut shells, and rice husks) in the humid tropics. Results showed that biochar from coffee husk were the most productive in the maize field. The soil pH in tropical area are so acidic (pH=4.7) and pH increasing because of soluble coffee husk biochar improve soil quality and efficiency for crop production.

4.6.4. Silage and composting. Silage is the direct usage of organic residues on soil surface without any treatment, while composting is the biological decomposition of organic waste promoted by bacteria, fungi, worms, and other organisms under controlled aerobic conditions to obtaine~~d~~ a partially decayed organic matter [115]. The chemical composition of coffee husk in terms on nutritive elements like N, K, P, and others (Table 2) makes it suitable to be used as amendment in agricultural soils.

4.6.4.1. Silage. Coffee husk silage can be a good option for K depleted soils, but there is the risk of phytotoxic production [116]. Application of raw coffee husk in the field inhibits the plants, specially the roots, while anaerobic decomposition increases the emission of greenhouse gases [30]. It was observed that addition of coffee husk on soil provided an increase in dry matter content, but also decreased the buffering capacity responsible for maintaining soil pH [15]. However, coffee husk spread at the soil surface may decrease the soil erosion, temperature, and evapotranspiration. So, notwithstanding the problems due to its phytotoxic activity, coffee husk can help in land reclamation [116].

4.6.4.2. Composting. One of the most important problem in coffee industry is by-products accumulation and, subsequently, economic and environmental costs for their management due to their potential contamination effect caused by the leaching of phenolic compounds. In fact, notwithstanding the many different uses to which coffee husk can be addressed, in coffee producer countries every year huge amounts of coffee husk are produced and, especially in developing countries, much of this husk is released in the land without any pre-treatment. Instead, phenolic acids content and mutagenic effects of caffeine require to treat coffee husk before land distribution to reduce its environmental concern. As reported at point 3.5.7, there are several ways to remove inhibitors from coffee husk, but composting is the most affordable, environmentally friendly, and efficient system. Because of this, different investigations aimed to improve waste management and ecosystem sustainability have been done on coffee husk so to transform a disposal problem into a valuable product for agriculture. Composting of coffee husk with other organic materials or alone is one of the best ways to profitably manage coffee husk since the process has capacity to solve management problems like mass accumulation and detoxification. Composting by oxygen-driven biological methods allows easily recycling great amounts of agricultural by-products and producing high-quality fertilizers [117][30]. Coffee husk has characteristics that make it suitable to be composted; for instance, it has a C/N ration around 30 [30] and is rich in lignocelluloses materials, which makes it an ideal substrate for microbial processes [2]. Inoculation of lignocellulosic waste materials with lignin-degrading microorganisms accelerates the composting process and improves compost quality and the humification process [118].

Dzung *et al.* [119] studied coffee husk supplemented with cow manure and lime. The mixture was composted for 3 months and then was supplemented with 0.1% (w/w) effective microorganisms like N_2-fixing *Azotobacter* sp. and *Bacillus megaterium*; the authors found that the quality of the obtained compost was better than some bio-organic fertilizers present on the agriculture market. This compost was applied on coffee field and the results showed that soil fertility, nutrient content in the coffee leaves, and the growth of the coffee plants were improved in comparison with the control. Sekhar *et al*. [120] applied different dosages of coffee husk compost with NPK fertilizers in various amounts in the paddy field and found that applications of 4 ton ha^{-1} of coffee husk compost plus 80 kg ha^{-1} N, 60 kg ha^{-1} P, and 50 kg ha^{-1} K gave the highest grain and straw yield production. Kassa and Workayehu [31] evaluated the quality of composts comparing the quality of only coffee husk compost with mixtures made of coffee husk+cow dung, coffee husk+*Millettia ferruginea*, coffee husk+cow dung+*Millettia ferruginea*, and coffee husk+effective microorganism, and concluded that the mixtures coffee husk+*Millettia ferruginea* and coffee husk+cow dung+*Millettia ferruginea* gave the highest quality composts. In the coffee husk composting experiments run by Bidappa [121][108] and Tuan [122], as we may deduce from the fact that the use of these composts improved soil fertility and crop yield, a strong reduction of phenolic compounds was obtained. Shemekite *et al.* [30] used cow dung and green wastes as co-substrates in the composting of coffee husk and monitored the physicochemical changes and the microbial community dynamics during the composting process. While at the beginning of the process the microbial communities of all the compost piles differed, they were similar at the end, as shown by DGGE fingerprints and microarray analysis.

Improving soil fertility and plant growth is one of the benefits coming from compost application in agriculture. Other helpful impacts are the decrease of soil erosion and evapotranspiration, which may contribute to land reclamation. Thus, since composting process disinfects organic wastes from pathogens and weed seeds and stabilized C, N, and other nutrients in the organic fraction, applying compost to the field can help to maintain or increase the soil organic matter content, biological activity, and porosity, so helping water, air, and plant roots to penetrate easily the soil [123][117].

4.7. RESUMING OF COFFEE HUSK APPLICATIONS ~~APPROACH~~

Table 9 shows a comprehensive view of the possible uses of coffee husk ~~as it comes from~~obtained by processing coffee ~~processing~~cherries by dry method.

Table 9. Possible uses of coffee husk in industrial, fuel, agri-food, and agriculture activities.

Application	Reference
Industrial use	
Ceramic	[81]
Particleboard	[26][82]
Flavor extraction	[83]
Fuel	
Solid fuel	[84][85]
Gasification	[86][87]
Ethanol production	[88][89][27]
Contaminants adsorption	
Lead (Pb)	[94][93]
Nickel (Ni)	[95]
cyanide	[96]
dye contaminants	[97]
norfloxacin	[98]
Fermented products	
Organic acid	[99][100]
Enzymes	[102]
Bioactive compounds	
Dietary fiber	[104]
Anthocyanin	[106]
Agriculture	
Animal food	[34]
Mushroom bed	[109]
Biochar	[112]
Silage	[116]
Compost	[30][120][121][122]

5. CONCLUSIONS

Coffee consumption in the world increases every year and the same happens for its by-products. Coffee husk is the main by-product of coffee roasting process by dry method and is one of the most abundant by-products that are spread in the land, giving rise to some environmental concerns. Nonetheless, coffee husk components make this material suitable to be used in several ways in many industrial, fuel, agri-food, and agriculture activities. Because of its high content of phenolic compounds, the use of coffee husk may require detoxification, and many systems have been identified to reduce the toxic effect of coffee husk; all these systems are reported in this review. The use of coffee husk as direct or indirect fuel is one of the most practiced way to recycle it but, because of its content in nutritive elements, the use in agriculture should be promoted, especially in acid soils, possibly after composting instead to be directly used as soil silage. However, the lack of local application and performing of the scientific results obtained at a global scale is a challenge that should be the topic of future studies in order to improve recycling of these valuable materials and increase soil fertility.

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