waste

Review

Spent Coffee Grounds Characterization and Reuse in Composting and Soil Amendment

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Abstract: As an everyday beverage, coffee is consumed worldwide, generating a high amount of waste after brewing, which needs attention for its disposal. These residues are referred to as spent coffee grounds (SCGs), which have been shown to have applications as polymers/composites precursors, biofuels, and biofertilizers. This review focuses on agricultural applications usually based on organic matter to fertilize the soil and consequently improve plant growth. To date, SCGs have been shown to exhibit outstanding performance when applied as soil amendment and composting because it is a nutrient-rich organic waste without heavy metals. Therefore, this review presents the different options to use SCGs in agriculture. First, SCG composition using different characterization techniques is presented to identify the main components. Then, a review is presented showing how SCG toxicity can be resolved when used alone in the soil, especially at high concentrations. In this case, SCG is shown to be effective not only to enhance plant growth, but also to enhance nutritional values without impacting the environment while substituting conventional fertilizers. Finally, a conclusion is presented with openings for future developments.

Keywords: spent coffee grounds; agriculture; biofertilizers; soil amendment; composting; characterization

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1. Introduction

For many years, coffee has been consumed all over the world. Today, coffee is a well-established beverage in various countries, Europe being the main coffee consumer market (54 million bags in 2020: 1 bag = 60 kg) [1,2]. It is also the second greatest traded commodity on the market, petroleum being the number one [3]. Worldwide, this huge market and demands have led to a high production rate of coffee of 169.6 million bags in 2020 [1]. Consequently, the coffee industry dramatically contributes to waste generation. Coffee pulp, coffee husks, coffee silverskin, and spent coffee grounds (SCGs) are the most important by-products of coffee processing (Figure 1) [4,5]. SCGs are produced from the treatment of coffee powder with hot water to prepare instant coffee. It is estimated that 1 ton of green coffee generates 650 kg of SCGs [6]. In addition, coffee residues contain organic materials which are reported to be highly pollutant, incurring severe environmental issues and contamination if discharged directly into the environment [7,8].

Environmental pollution problems due to solid waste and new environmental regulations have forced the industry to pay more attention to waste management. SCGs contain valuable organic compounds (fatty acids, proteins, cellulose, and lignin); therefore, this large amount of waste can be considered as an excellent candidate not only to produce clean energy and value-added compounds [9–11], but also to be used as bio-fertilizers [2,12,13]. Recent studies showed that SCGs have excellent potential to be used in agriculture because of their high amount of nutrients and low risk of heavy metal contamination [14,15]. Therefore, SCGs have already been reported to be a soil amendment or a top dressing to improve soil quality and control weeds [12,16,17], as well as composting with animal manure and other wastes to improve soil quality, enhance plant growth, and avoid intoxicating the soil [18,19].

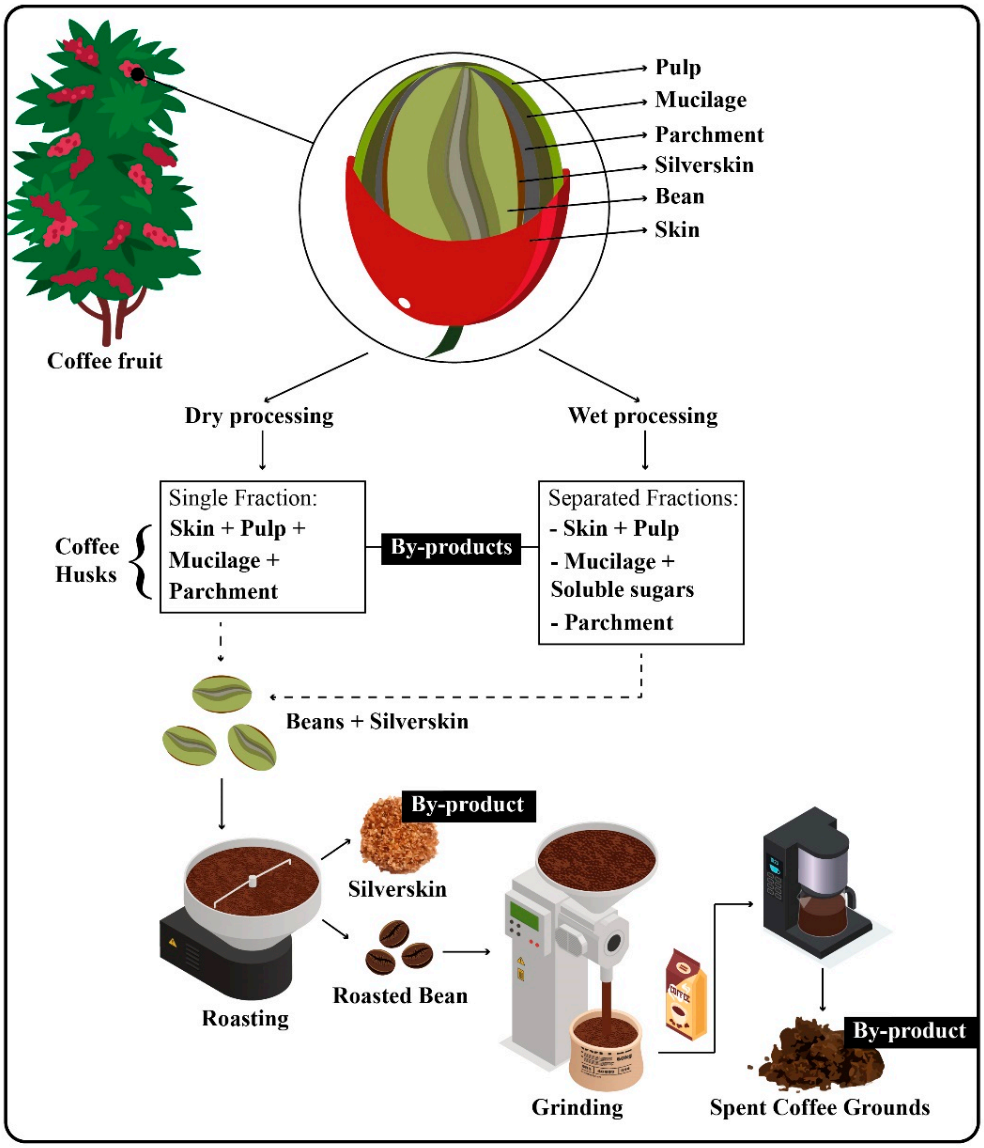

Figure 1. Coffee production and by-products.

This review focuses on SCG characterization, including its physicochemical composition and the techniques used for these investigations. Then, studies on SCGs in agriculture as soil amendment and composting are discussed, while recommendations are provided for future studies. Although other reviews have been published on the valorization of SCG in several fields [6,10,20], none of them focused on SCGs as a biofertilizer for agriculture. Therefore, this review provides valuable information to guide future research efforts and regulations regarding the valorization of SCGs in the agriculture field.

2. SCG Composition

Carbohydrates, lignins, lipids, minerals, and proteins are the main components found in the chemical composition of SCG (Table 1) [21]. It is also possible to find phenolic compounds (12.0 mg/g), caffeine (14.5 µg/g), and chlorogenic acid (31.8 µg/g) [13]. Carbohydrates (polysaccharides), such as cellulose and hemicelluloses, are highly present in SCGs. Cellulose, in general, is a linear homopolymer of repeated glucose units leading to potential pulp and paper production applications. Moreover, because cellulose can be converted to glucose (sugar), cellulose from SCG can be subsequently used in ethanol and butanol production [9]. Hemicelluloses, as heteropolymers, can release different types of sugars through chemical or biological pre-treatments. Therefore, mannose, arabinose, and galactose are typical examples of sugars obtained from hemicelluloses [22]. It was reported that polysaccharides can be extracted from SCGs by alkali treatment, resulting in 39.0% of total sugar, with different monosaccharide yields in the form of arabinose (19.9% mol), mannose (4.4% mol), galactose (60.3% mol), and glucose (15.4% mol) [23]. A more recent study identified a carbohydrate content of 67.4 wt.% (dry mass) and a monosaccharide composition of arabinose (12.0% mol), mannose (48.4% mol), galactose (22.5% mol), and glucose (17.1% mol) [24]. The authors also performed an alkaline peroxide (35% v/v) treatment on SCG and observed that the carbohydrate content decreased to 64.2 wt.%.

SCGs have a high protein content (13–17 wt.% of dry mass) [9,24]. Hence, they can be used as a reliable nutrient supply in the food industry and potentially play an important role as substrates in biotechnological and fermentative processes [20]. Recently, SCG proteins were extracted by a urea-based method followed by hydrolysis with two different enzymes (alcalase and thermolysin) to produce bioactive peptides. The results showed peptide yields of 0.23 mg/mL (alcalase) and 0.39 mg/mL (thermolysin) after polyphenol extraction, as well as antioxidant activities (hydroxyl radical assay) of 84.3% and 95.4%, respectively [25]. In another study, SCG proteins (58.6%) were shown to exhibit antihypertensive and antioxidant potentials, leading to potential applications in human nutrition for glycemic and hypertense control [26].

Numerous studies also explored the extraction of value-added products from lipid fractions, such as coffee oil (7–21% of total SCG dry mass) [20,27,28]. This oil has been used for biopolymer [29], biofuel [30], and sunscreen [31] production. Phenolic compounds have been extracted from coffee oil and studied due to their antioxidant potential. They are being used in functional and supplement food, as well as in seed germination [32,33]. One study identified 230.2 mg GAE/g (GAE = µmol gallic acid equivalents) of phenolic compounds in alkali-pretreated SCG and 234.2 mg GAE/g in SCG hydrolysates, whereas another study identified 5.7 mg GAE/g of phenolic compounds in SCG extracts [33,34]. Moreover, it was proposed an environmentally friendly method to extract SCG phenolic compounds via ethanol–solvent extraction and the authors reported over 90% recovery of the phenolic compounds from SCGs [32].

Minerals are also found in the SCG chemical composition. For this reason, compost substrates based on SCGs can help to limit the extraction of minerals from the soil [35]. According to Table 1, potassium, phosphorus, magnesium, and calcium are the most abundant minerals extracted from SCGs.

Table 1. Chemical compounds and minerals compounds found in SCGs.

Chemical Compounds	Content (wt.%) [1]	References	Minerals Compounds	Content [2] (mg/g) [1]	References
Hemicellulose	30–40	[9,20,36]	Potassium	3.70	[22,37–39]
Cellulose	8–15		Phosphorus	1.47	
Lignin	20–30		Magnesium	1.29	
Proteins	13–17		Calcium	1.38	
Lipids	7–21		Sodium	0.07	
Ashes	1–2		Aluminum	0.28	
			Iron	0.12	
			Manganese	0.05	
			Copper	0.03	
			Zinc	0.01	

[1] Dry weight. [2] Only the highest values are reported.

This wide variety of nutrient-rich compounds extracted from SCG can lead to various applications depending on the composition, especially in agriculture as fertilizers. Consequently, the characterization of these residues plays an essential role in determining the most appropriate applications.

3. SCG Characterization Techniques

This section is divided by subheadings. It provides a concise and precise description of the experimental results, their interpretation, as well as the experimental conclusions that can be drawn. The characterization of SCG is essential to determine their appropriate use and to select potential applications and treatments [40]. Several rapid and cost-effective techniques have been proposed and implemented to determine the physicochemical properties of these natural materials (Figure 2).

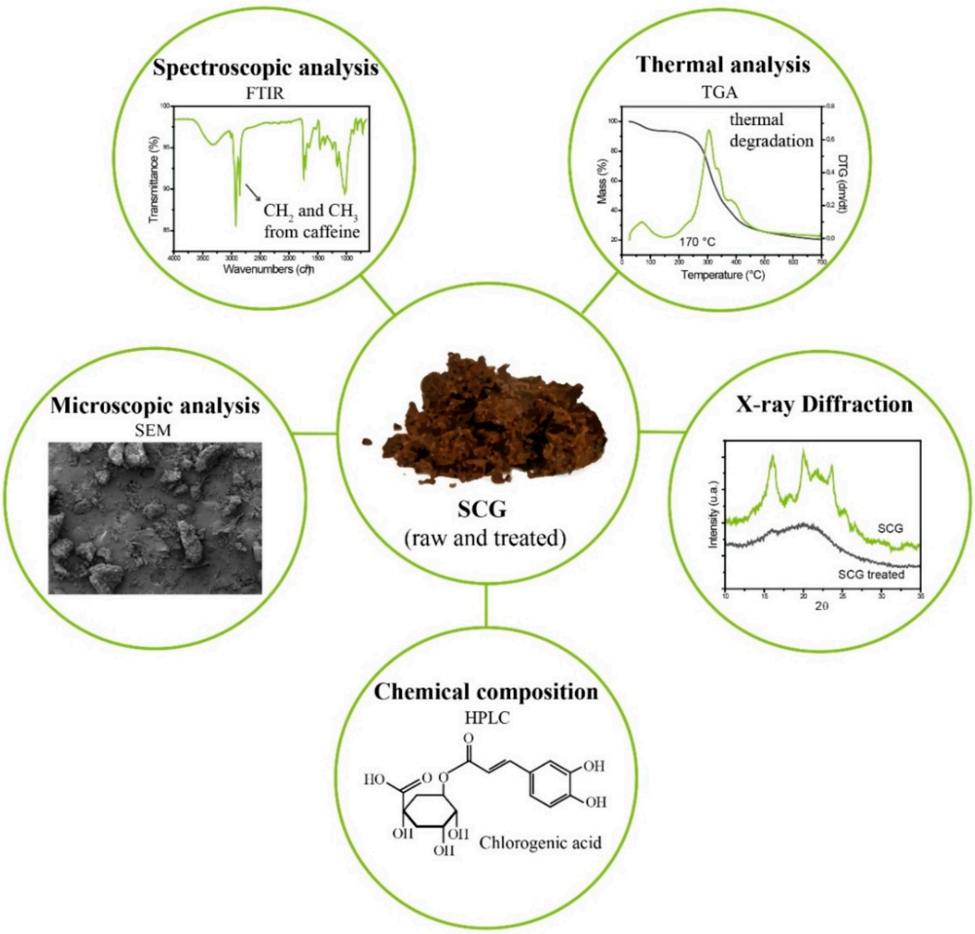

Figure 2. Typical SCG characterization techniques.

3.1. Spectroscopic Analysis

Spectroscopic techniques are measurements related to the absorption, emission, or scattering of electromagnetic radiation by atoms or molecules [41]. Infrared (IR), as a well-known spectroscopic technique, has been widely used in the quality control of different foods, especially coffee products. The IR spectrum is divided into three regions: far-infrared (10–400 cm^{-1}), mid-infrared (400–4000 cm^{-1}), and near-infrared (4000–14,000 cm^{-1}) [41,42]. It should be noted that energy levels related to mid-infrared (MIR) and near-infrared (NIR) regions are not adequate to excite electrons, although they can induce vibrational excitation of covalently bonded atoms and groups. Each functional group or bond between atoms presents a unique vibrational frequency; therefore, they can specify the chemical bonds included in a sample [41,43].

A recent study used NIR spectroscopy to determine coffee flavors in coffee beverages through machine learning and deep learning models, where seven different flavors were effectively predicted in 266 samples [44]. Another study used NIR with PLS to identify the moisture content and soluble solids, as well as the total and reducing sugars of Brazilian green coffee. Results showed that moisture content provided the best statistics results. Water absorption bands are located at 970, 1450, and 1940 nm, for which the 1940 nm band was the best to characterize the moisture content, with OH group vibrations and elongations [45].

NIR was also used to identify three phenolic compounds in SCGs—caffeic acid, (+)-catechin, and chlorogenic acid—as well as three methylxanthines compounds—caffeine, theobromine, and theophylline [46]. Furthermore, it was applied an online NIR method to predict the acidity during the coffee roasting process, estimated to be 0.16 mL NaOH/g. The proposed methodology was developed based on different coffee varieties (Arabica and Robusta), coffee origins (Brazil, East-Timor, India, and Uganda), and roasting process (slow and fast) [47]. In addition, diffuse reflectance infrared transform spectroscopy (DRITS) was effectively used to discriminate roasted coffee, SCG, and common adulterants such as coffee husks and corn [48,49].

Fourier-transform infrared spectroscopy (FTIR) was performed to discriminate between defective and non-defective coffee beans using the multivariate statistical analysis technique (MA) [50]. FTIR was also used to perform qualitative and quantitative analysis on Kona coffee grounds, brewed and their blends. The authors were able to predict the Kona coffee content inside the blends [51]. An FTIR/PCA system integrated with chemometrics was used to classify unadulterated and adulterated coffee samples. It is worth mentioning that successful classification of the samples based on the sibutramine content in green tea, green coffee, and mixed herbal tea was achieved with perfect accuracy, without false prediction [52]. Another study used FTIR to identify green coffee beans from six different origins. The spectroscopic curves were calibrated based on the main coffee compounds (lipids, proteins, and carbohydrates), enabling adequate identification of symmetric and asymmetric CH_2 vibrations, as well as ester (C=O) vibrations [53].

In addition to infrared, other techniques have been carried out. Nuclear magnetic resonance (NMR) spectroscopy was applied for coffee Arabica samples from America, Africa, and Asia to identify fatty acids as the main compounds in American samples, chlorogenic acids and lactate in African samples, and acetate and trigonelline in Asian samples [54]. Additionally, NMR spectroscopy was used to identify that SCGs are an abundant source of lipids and fatty acids, as are roasted coffee beans [55]. UV–visible spectroscopy was also reported with different approaches for coffee characterization, such as to classify Java Island coffees, for which the authors found a specific identification (marker) for each coffee type in the 250–450 nm wavelength range [56]. Moreover, Raman spectroscopy was used to confirm the obtention of carbon-based materials from SCGs (via catalytic carbonization), which was confirmed by the formation of graphitic carbon in the samples. The graphitization level was quantified by the intensity of the peaks at 1345 cm^{-1} (D band) and 1581 cm^{-1} (G band) [57]. The same behavior was observed in heavy metal-free carbon obtained from SCGs using KOH-urea and NaOH-urea as activators [58].

Spectroscopic techniques have also been reported in SCG composting for fertilizers to identify the compost maturity [19,59] and organic matter humification [60]. SCG applications as agriculture fertilizers are discussed in Section 4.

3.2. Thermal Analysis

In terms of lignocellulosic materials, thermogravimetric analysis (TGA) is the most commonly performed thermal analysis technique, which usually focuses on identifying the thermal decomposition and the mass changes of these organic materials for different applications. TGA measures the material's mass as a function of temperature or time, while this material is subjected to a controlled temperature under a specified atmosphere (usually air or nitrogen) [61].

TGA was coupled with a mass spectrometer (MS) to identify the SCG thermochemical decomposition [62]. The study showed that it was possible to conduct pyrolysis tests under different heating rates (5, 10, 15, 25, 50, and 100 °C/min) up to 500 °C. It was found that the heating rate had a significant effect on the SCG thermal decomposition because the decomposition rate linearly increased with increasing heating rate. Another study reported that TGA via mass differences can determine the fatty acids content in lipids extracted from SCG and commercial lipids [63]. Polysaccharides from SCG were also analyzed by TGA, and the results showed that these compounds have similar thermal characteristics to well-known polysaccharides (including cellulose and hemicellulose) [64]. TGA was combined with DSC to analyze polysaccharides from SCG [23] and to compare SCG with coffee silverskin [36]. All these studies were able to identify the thermal decomposition (300 °C) and weight loss (around 40%) of SCG and the polysaccharides isolated from SCG. Additionally, DSC showed one endothermic event (around 75 °C) corresponding to water vaporization (also found in TGA), and one exothermic event (around 310 °C) corresponding to the thermal depolymerization and the branching of SCG and SCG polysaccharides [23,36,65]. Another authors also worked with TGA and DSC, focusing on the thermal stability of SCG after extraction of the oil fraction, where the oil was used to produce a high-quality and stable biodiesel, while the remaining SCGs went to compost [66].

SCG films with pectin [67], cellulose [68], and galactomannans [24] were successfully evaluated by TGA to compare the thermal properties of the films with neat SCG for packaging applications. Thermal stability and mechanical strength results of cellulose with alkali-treated SCG showed that this film could be used for packaging photo-sensitive materials, such as flowers and vegetables [68]. In addition, aerogels based on SCGs and cotton cellulose were developed for which the thermal stability and thermal conductivity were analyzed by TGA and a with a C-Therm TCi Thermal Conductivity Analyzer (TCA). This study confirmed the aerogels' thermal properties for thermal insulation applications [69].

Several studies have reported SCGs as a filler for polymer composite production. Thermal analysis is one of the main characterizations to perform, because the processing temperature is generally relatively high (above Tm for thermoplastic resins) to ensure limited thermal decomposition of the lignocellulosic filler. For example, different matrices can be filled with SCGs, such as polypropylene (PP) [70–72], polyethylene (PE) [73,74], polylactic acid (PLA) [75,76], polyurethane (PU) [77], poly(vinyl) alcohol (PVOH) [78], poly(butylene adipate-co-terephthalate) (PBAT) [79,80], and epoxy (EP) [81]. More information on the subject can be found in our previous review [11]. Most of these studies performed TGA prior to composite production and compared the thermal properties of the raw materials with the final composites.

TGA was also combined with simultaneous differential thermal analysis (SDTA), in which the analyses were carried out simultaneously under the same heating rate, to identify the thermal stability index (calculated by dividing the first soil mass loss by the total of both soils' mass loss) and the thermal decomposition of two different Mediterranean soils mixed with SCG (2.5 and 10 wt.%). The authors concluded that 10 wt.% SCG could decrease the thermostability index, indicating a less stable organic matter being more accessible to be decomposed by microorganisms [12].

3.3. X-ray Diffraction Analysis (XRD)

XRD analysis is widely used to measure the crystallinity of lignocellulosic materials. SCGs, as semi-crystalline material, has crystalline and amorphous regions. Cellulose is crystalline, whereas hemicellulose and lignins are amorphous [82]. In an XRD pattern, the peak at 11–12° was reported to correspond to the amorphous region, whereas the 21–22° range corresponds to the crystalline region, mainly from cellulose [68,83]. A comparative study was reported among the XRD patterns from SCG cellulose and SCG cellulose nanocrystal [83]. The results showed three peaks at 11.9°, 21.4°, and 22.7° for SCG cellulose. In contrast, SCG cellulose nanocrystal did not present the 11.9° peak, which indicates the efficiency of the acid hydrolysis treatment to produce cellulose nanocrystal by removing the amorphous components. The peak at 21.4° was used to calculate the crystallinity, leading to a value of 72%. Another study compared SCG with coffee silverskin, showing that the latter had more cellulose than SCG (23.7 vs. 12.4 g/100 g dry mass) [36]. However, the XRD pattern showed that SCG was more crystalline than coffee silverskin, which was related to the difference between the cellulose structure of both materials associated with the brewing process that SCG underwent (high pressure, temperature, and humidity).

Concerning biofilms, treated SCGs (0.05, 0.1, and 0.2 *w/v*%) were mixed with carboxymethyl cellulose (CMC) to improve the antioxidant and antimicrobial properties of the biofilm for food applications. XRD results showed that alkali-treated SCGs and autohydrolyzed SCG improved the crystallinity of the biofilm compared with the neat CMC biofilm, which was related to molecular chain mobility [34]. Another study identified that the addition of torrefied SCG (10–30 wt.%) in PBAT biofilms reduced the crystalline peak (22.9°) as the filler content increased [80]. XRD analysis also showed that alkali-treated SCG improved the crystallinity as compared with untreated SCG (from 30.6% to 34.4%), whereas enzymatically treated SCG slightly decreased the crystallinity (29.3%). However, mechanical tests showed that both SCG treatments exhibited similar tensile strength when applied to galactomannans biofilms, but enzymatically treated SCGs presented less deformation [24].

It was reported that SCG dispersion in a polymer matrix could reorganize the crystallographic planes inside the matrix because these particles reduce chain mobility of the polymer, acting as an impurity, and consequently leading to changes in the composite's crystallinity compared with the neat polymer. For example, high-density polyethylene (HDPE) filled with SCGs (10–30 wt.%) had a crystallinity of 84% for the neat polymer; the value decreased to 81% for the composite [74]. In another study, poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) filled with SCGs (5 and 25 wt.%) presented 17.4% and 9.7% crystallinity, whereas the neat PHBV (32.2%) and SCG (21.7%) had much higher values [84].

3.4. Chemical Composition

The chemical composition of SCG was determined by various methods and techniques depending on the objective of the analysis, i.e., the specific compounds to be quantified. High-performance liquid chromatography (HPLC) is widely reported to quantify polysaccharides (cellulose and hemicellulose), monosaccharides (glucose and mannose), phenolic compounds (caffeine), and organic acids (acetic acid and lactic acid) [22,30,36,64,85,86]. One of these studies measured the sugars, glycerol, ethanol, organic acids, phenolic acids, and alkaloids by HPLC in SCG hydrolysates before and after fermentation [85]. The results showed that after fermentation, an increase in organic acids content, such as succinic acid (from 2.67 to 14.86 g/L) and lactic acid (from 0.12 to 18.46 g/L), was observed, and a decrease in the sugars content, such as glucose (from 52.68 to 4.15 g/L) and fructose (from 18.70 to 2.64 g/L), occurred.

Monosaccharides and phenolic compounds of SCGs were also determined by gas chromatography (GC) [24,87,88]. Phenolic compounds extracted from SCG (wet mass) were identified as 0.38 g/g, representing 13% of the total oil fraction from SCGs [9]. Another study measured and compared the number of monosaccharides in untreated and alkali-treated SCGs, such as arabinose (12.0 and 3.9% mol), mannose (48.3 and 53.9% mol), galactose (22.5 and 28.8% mol), and glucose (17.1 and 13.3% mol) [24]. Additionally, the total nitrogen and protein contents were measured via the micro-Kjeldahl method [87,89,90], and the lipid content was measured via Soxhlet extraction [24,36]. Proteins of SCGs (10.9%) and coffee beans (11.6%) were measured, and the results showed that the proteins from coffee beans could positively influence the adsorption of lead and iron from drinking water [89].

SCG nutrients, such as calcium (1.5 g/kg), magnesium (19.0 g/kg), potassium (2.9 g/kg), sodium (4.5 g/kg), iron (0.05 g/kg), zinc (0.009 g/kg), manganese (0.03 g/kg), and copper (0.02 g/kg), were measured by atomic absorption spectrometry (AAS) to investigate the use of SCGs and tea leaf nutrients to improve the soil quality for rice crops [39]. In another study, calcium, manganese, phosphorus, and potassium contents were measured in SCG composting (combined with fungi activator and temperature control) by inductively coupled plasma optical emission spectrometry (ICP-OES). The results showed that after 28 days, the quality of SCG composting was improved, presenting an increase in all nutrients compared with the initial sample, including phosphorus (3.3 mg/g to 5.7 mg/g) and calcium (9.7 mg/g to 21.2 mg/g) [19]. The main nutrients and chemical compounds of SCGs are reported in Table 1.

3.5. Morphological Analyses

The morphology of SCGs was determined via scanning electron microscopy (SEM). Through SEM analysis, Bejenari et al. (2021) measured the average size of SCG particles (from 297 to 430 µm) and the pore size (from 20 to 30 µm) [91]. Nevertheless, Arrigo et al. (2019) showed that biochar produced from SCGs presented a sponge-like morphology with a lower pore size between 10 and 20 µm compared with untreated SCG [73].

In other studies, the SCG morphological structure was compared with coffee silverskin, coffee chaff, and hydrochar from SCG and SCG after oil removal. Coffee silverskin presented a denser morphology than SCG [36], whereas coffee chaff presented a fibrous morphology with a dense and smooth surface, which was different from SCG particles, having a porous and granular morphology associated with the removal of some components after brewing [92]. On the other hand, hydrochar from SCG showed a higher porous surface than neat SCG due to the hydrolysis process [93], whereas the SCG morphology after oil extraction was very smooth [94].

SEM analysis was also used to characterize SCGs as a filler for polymer composites (dispersion), as well as to characterize the adhesion between the matrix and the filler [71,74,95]. It was stated that composites filled with SCGs should not be called composites reinforced with natural fibers, but composites reinforced with natural particles because SCG has more of a particle shape and not a fiber shape (size less than 200 µm) [96]. Other authors investigated the morphology of SCGs with and without chemical treatment (bleaching), concluding that removing some components caused micropore formation, leading to a high porosity that can improve the adhesion/dispersion in a polymer matrix [97].

4. SCGs for Agriculture

SCGs have been studied as a good source for agricultural applications exploring more sustainable practices. SCGs contain large amounts of organic compounds, such as fatty acids, polyphenols, minerals, and polysaccharides; therefore, they can be exploited as a source of nutrients. One of the advantages of using SCGs as fertilizers is that SCGs do not contain heavy metals, which may be hazardous to the food chain [14,98]. In fact, it was reported that biochar from SCGs could be used to amend or adsorb heavy metals in the soil [99]. SCG can also reduce the mobility of heavy metals [100]. It is worth mentioning that crops grown with fertilizers based on SCGs are categorized as organic products in the European Union [14]. The following sections will briefly review various studies on soil nutrient enhancement through SCG focusing on soil amendment and composting. Furthermore, Table 2 shows the principal physicochemical properties of SCGs used as a soil amendment and in composting.

Table 2. Main physicochemical properties for SCG applications as soil amendment and composting.

Properties	Samples					
	SCG	**SCG as Soil Amendment [1] (7.5–15 wt.%)**	**SCG Hydrochar as Soil Amendment [2]**	**SCG and Cat Manure [3] (3:1 wt.%)**	**Vermicompost and SCG [4]**	**SCG and Chicken Eggshell [5] (200:20 g)**
pH	5.0–6.0	7.9	8.1	8.2	7.2	5.9
Moisture (%)	7.0–8.0	-	-	2.6	-	-
C (%)	47.0–60.0	-	0.0008	-	45.0	46.6
N (%)	1.8–2.5	0.07	0.00007	7.0	2.98	2.36
C/N	21–32	-	11	-	15.10	19.7
K (mg/g)	3.70	1.41	1.82	5.4	3.80	4.99
P (mg/g)	1.47	0.10	0.05	1.2	11.10	1.49
Mg (mg/g)	1.29	0.13	0.21	-	1.60	1.76
Ca (mg/g)	1.38	0.56	2.97	-	2.00	35.98
Na (mg/g)	0.07	-	0.48	-	0.80	0.20
Fe (mg/g)	0.12	0.005	1.22	-	-	-
Mn (mg/g)	0.05	0.003	0.04	-	0.06	-
Cu (mg/g)	0.03	-	0.003	0.004	0.03	-
Zn (mg/g)	0.01	-	0.01	1.2	0.02	-
References	[12,17,37–39,93,101]	[15]	[17]	[100]	[38]	[18]

[1] Values considering lettuce production after 40 days of cultivation. [2] Values considering hydrochar from raw SCG applied to topsoil after 28 days. [3] Values considering the composting applied to soil after 31 days of incubation. [4] Values after 72 days of composting. [5] Values after 3 months of incubation.

4.1. Soil Amendment

SCG may be used as soil amendment or mulch. When used as mulch, it mitigates soil temperature and retains water in the soil due to the high water-holding capacity, similarly to any other mulch materials [36]. SCGs also bind pesticide residues or other toxic heavy metals in the soils and inhibit their movement [99]. The most significant effect of SCG in soil is to increase the availability of essential soil nutrients for various domestic plants.

SCG was investigated and applied to soil organic matter of two Mediterranean agriculture soils, vega soil (58% of clay) and red soil (43% of clay), for a short period (30 and 60 days). The results showed improvements in both soil organic matters with 2.5% and 10% of SCG. However, vega soil provided a better organic matter fraction (16,964 mg C/kg vs. 18,382 mg C/kg), which was associated with its higher clay content (58%) and higher carbon content (1.4%), generating a higher-quality soil. Nevertheless, the authors claimed that the quality analysis of organic matter should be performed more extensively [12].

Incubation and mixing with espresso SCGs and soil in field experiments was conducted to determine the effect of SCGs on lettuce, carrot, and spinach growth. They studied the mineralization and immobilization of nitrogen in the soil over time, and showed that SCGs had no positive effect on lettuce, spinach, and carrot growth, nor improved their nitrogen supply. This effect was due to the presence of caffeine having an inhibitory effect on plant growth [102]. Caffeine is a natural secondary metabolite with biological roles in some plants, such as coffee and tea, serving as a chemical defense mechanism. It is also believed to inhibit germination [103]. Other studies reported this issue to be a decisive parameter in applying SCGs in agriculture [104–107].

In another experiment, the authors studied the effect of SCGs on chlorophyll and carotenoid contained in lettuce with the addition of 0% to 20% (*v*/*v*) fresh SCGs to the soil. They found that chlorophyll was increased by up to 37%, while organic nitrogen decreased by only 4.4% at the optimum SCG content of 10%. Therefore, SCG addition can be effective at low contents (up to 10%), but the nitrogen supply tends to decrease even more at higher SCG content (20%) because only mineralized nitrogen is absorbed by plant roots (NO_3^- or NH_4^+), whereas the SCG nitrogen content is in the form of organic compounds such as proteins, melanoidins, and caffeine [108]. A more recent study reported a significant reduction (35% reduction with 15% SCG) in the soil nitrogen content for lettuce production [15]. However, the authors highlighted the improvement in the lettuce's nutritional elements with the presence of SCGs in the soil: Fe (2.0 to 5.4 μm/g), Ca (489 to

563 μm/g), and Mn (1.4 to 3.1 μm/g) contents increased. The same authors evaluated the use of fresh SCG and hydrolyzed SCG, as well as SCG washed with water and ethanol for composting, vermicomposting, and biochar for lettuce production [109]. They concluded that SCG as a vermicomposting feedstock and biochar are the best options for lettuce growth, because increased nitrogen contents of 260% (vermicomposting) and 63% (biochar) were observed without intoxicating the soil, because no phenolic compounds, such as caffeine, were present. Nevertheless, vermicomposting and biochar provided a poorer nutrient soil than fresh SCG. In a third study, the authors investigated the use of washed hydrochar from SCGs (second-generation coffee waste) to improve lettuce production. The authors reported that hydrochar has better physicochemical properties than SCGs in terms of the carbon/nitrogen (C/N) ratio (29 and 24, respectively), organic carbon (56% and 47%, respectively), and polyphenols (186 and 77 mg GAE/g, respectively), but they both presented similar properties when applied to the soil. Nevertheless, because of the amount of polyphenol generated after the hydrothermal carbonization process, hydrochar also inhibited lettuce growth. Despite this issue, the biochar generated a higher nutrient content. For example, 2.5% of hydrochar at 185 °C increased the Ca (141 to 168 mg/100 g), Mn (0.28 to 0.42 mg/100 g), and Fe (0.66 to 1.73 mg/100 g) contents [93].

SCG was also incorporated with horse manure into the soil and found that the addition of 10 kg/m^2 of SCG provided an increase of 25% in carbon content and 45% in nitrogen content, resulting in a reduction in the C/N ratio (from 13 to 10) as compared with the addition of horse manure alone. The authors also explained that the high nitrogen content in the soil resulted from the accumulation of insoluble nitrogen. Top-dressing of SCGs was also tested, but this was ineffective and did not induce any decrease in the C/N ratio, allowing weed control for only six months [110]. Moreover, a study was performed with cat manure and SCGs to eliminate cat manure toxicity in spinach crop production. SCGs were efficient in enhancing spinach growth and preventing toxic elements in the soil, but did not improve the soil nutrient contents (Zn, Cu, Cd, and Pb) compared with cat manure alone: SCG/cat manure presented contents of Zn = 1.39 mg/kg and Cu = 0.90 mg/kg, whereas cat manure alone presented Zn = 23.53 mg/kg and Cu = 8.73 mg/kg considering the root of spinach (dry weight) [100].

Iron is an essential nutrient for the soil, carrying essential elements through the plant's circulatory system and improving chlorophyll production. In an experimental study, the authors claimed that SCGs and tea wastes contain substances that could be used as Fe-chelating agents to enhance the required Fe of plants in the soil. They conducted incubation and pot experiments on two test soils and reported that the application of 40 μg/g of Fe in calcareous soil caused a significant increase (more than 100%) in the Fe values for soil treated with SCGs and tea wastes after 60 days as compared with those treated with ferrous sulfate ($FeSO_4$) alone [111]. Eliminating Fe deficiencies in farm-scale production is an important economic issue; therefore, this study has suggested a compelling solution using SCGs and tea wastes. Following this research, another study was conducted on improving rice grains' mineral content (especially Fe and Zn). They designed a series of experiments to directly compare the effect of the top-dressing application of ferrous sulfate ($FeSO_4$) and zinc sulfate ($ZnSO_4$) or Fe and Zn enriched with coffee or tea waste on rice paddy fields. They reported that brown rice's highest Fe and Zn concentrations were found while applying enriched coffee and tea waste. This was attributed to Fe(II) precipitation in the rice rhizosphere [39]. Accordingly, enriching coffee wastes with Fe and Zn seems to be an effective application to rice fields. It is worth mentioning that higher Fe and Zn concentrations in rice were related to the metal chelating capacity of organic materials, such as coffee and tea wastes, as previously confirmed by other studies [33,112].

For wheat–soybean crops, SCGs were successfully used as a top dressing in the soil after crop germination. The results showed that 10 kg/m of SCG enhanced weed control, reducing growth by 50%, whereas 5 kg/m of SCG improved the C and N contents [16]. Moreover, a recent study concluded that applying a pre-treatment on SCG, such as hydrothermal carbonization and alkali treatment, could also solve the toxicity effect of SCG components, as well as improve plant growth by increasing the rosette diameter, the number of new leaves, and the dry weight compared with untreated SCG [17]. Another study investigated the effect of SCG by-products in soil amendment, considering C and N values, such as biochar at 270 and 400 °C, hydrochar at 160 and 200 °C, defatted SCG, biochar from defatted SCG at 270 and 400 °C, and vermicompost [101]. Hydrochar samples presented N immobilization and lower maintenance of C in the soil, whereas the biochar presented a lower N immobilization and better maintenance of C in the soil, although vermicompost did not present N immobilization. The authors concluded that further analyses should be carried out on soil quality and fertility.

Experiments were conducted to study fresh and composted SCGs as fertilizers for lettuce growth in domestic agriculture. They reported that at low concentrations ($\leq$2.5 wt.%) of fresh SCGs and high concentrations (15 wt.%) of composted SCGs, the lettuce growth rate was substantially improved (up to 100% for fresh SCG at 2.5 wt.%, and up to 55% for compost SCG at 15 wt.%). They claimed that high fresh SCG content induced plant stress due to the presence of caffeine, whereas composted SCGs contain fewer toxic components [7]. These results showed that composting could be an alternative to reducing SCG toxicity, which has been extensively studied over recent years.

4.2. Composting

Composting is defined as the biological decomposition of organic waste material by bacteria, fungi, worms, or other organisms under controlled aerobic conditions [59,113] (Figure 2). The aerobic microorganisms involved in composting use oxygen to convert the organic matter into heat, water, CO_2, NH_3, and humus (decayed organic matter). In other words, the composting process converts the organic matter to a stable product called humus by the action of microorganisms with proper temperature and moisture and, consequently, reduces the organic waste volume in landfills [59,114]. Organic waste makes up a significant fraction of solid waste in some parts of the world; therefore, composting is gradually developing as an alternative to landfilling [115–117].

Successful composting requires the optimization of several parameters depending on the formulation of the composting mix, such as bulk density, porosity, particle size, nutrient content, and C/N ratio, as well as several process parameters such as temperature, pH, moisture, and oxygen supply [116]. The C/N ratio is the main factor related to compost maturity, where a high C/N ratio leads to a prolonged process, with carbon and nitrogen quickly metabolized. On the other hand, there is an excess of nitrogen per degradable organic matter in the case of a low C/N ratio, which can be lost by ammonia volatilization [59]. Therefore, an optimal C/N ratio is in the range of 25–35 [59,60]. In fact, the microorganisms require an energy source, such as organic carbon and nitrogen, for their development and activity. The pH was not believed to be an important parameter in the composting process, because most of the organic wastes are within the recommended range of 5.5–9 [60]. However, a pH < 7.5 can be better to control nitrogen losses caused by ammonia volatilization during composting with animals' manure. The microorganisms used in composting are from a vast group of microbial populations, and they develop according to the temperature of the organic matter (the processing steps). In the early steps, 50–60% of water content is desirable, while the optimum temperature is 40–65 °C [60].

Among various composting technologies, vermicomposting is seen as a sustainable technique involving earthworms as a natural reactor for converting solid waste into vermicompost or vermicast, reported to be a pollution control bioprocess [118]. In fact, it is a cooperation between microorganisms and earthworms, in which the former are responsible for the biochemical degradation of organic wastes, while the latter treat the decomposed matter and alter the biological activity, accelerating the composting process [119]. Vermicompost is believed to contain hormones and enzymes stimulating plant growth, produced during organic matter processing in the earthworm body [118]. The main tasks of earthworms in vermicomposting include substrate aeration, mixing, grinding, fragmentation, and enzymatic digestion, as well as microbial decomposition of the substrate in their intestines [120,121].

SCGs have been mixed with other organic wastes to produce a rich organic matter for agriculture fields, reducing toxicity when used alone as a fertilizer (Figure 3). The first published research in the vermicomposting area was from 2009 [122]. This pioneering study used an earthworm *Lumbricus rubellus* with three different composts comprising cow dung, kitchen waste, and SCGs. They claimed that the fine grinding of SCGs had several advantages, such as improving the compost texture enhancing the water retention capability, facilitating earthworm reproduction by stabilizing the pH due to increased air circulation, and reducing the temperature to an appropriate range for reproduction, as well as preventing pests from disturbing the earthworms due to the coffee aroma [122,123]. For the nutrient content, SCG addition to cow dung and kitchen waste (35:30:35) increased the nitrogen (~96%), potassium (~100%), and magnesium (~40%) contents compared with the mixed cow dung and kitchen waste (30:70). Although the nitrogen increased, the organic carbon presented similar trends, leading to a reduction in the C/N ratio from 14.1 (cow dung with kitchen waste) to 7.1 for SCG with cow dung and kitchen waste [122].

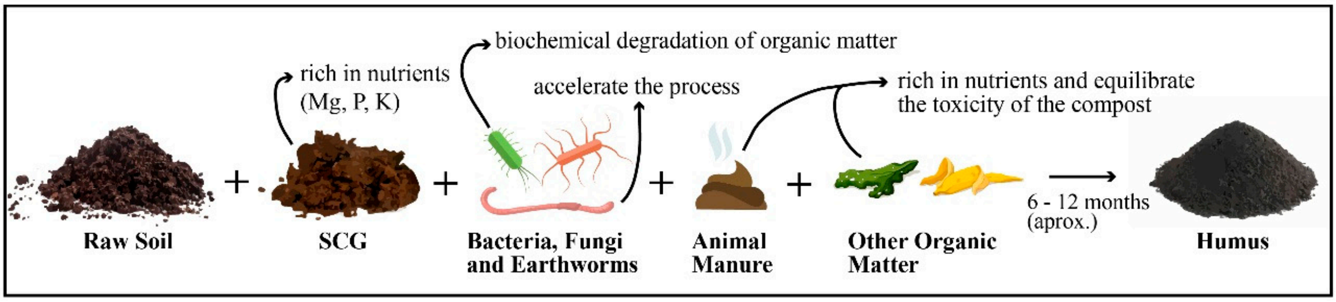

Figure 3. Composting scheme with SCG and other commonly used elements.

Another work compared and evaluated in-vessel composting, vermicomposting, and aerated static composting using SCGs and SCGs mixed with coffee filters and waste cardboard to assess the nutrient contents and decomposition rates. For nutrient concentrations, aerated static composting presented phosphorous and potassium concentrations at least 1.5 times higher as compared with the initial nutrient content. This increase in phosphorus and potassium content (non-volatile compounds) is related to the mass loss of carbon and volatile compounds during the composting process (decomposition of organic matter). All three studied composts led to an increase in total nitrogen and a decrease in total carbon, for which the aerated static composting samples provided the highest nitrogen increase (75%), whereas cardboard samples presented a reduction in the nitrogen content [38]. Moreover, keeping nitrogen in compost to release it to the plant has always been challenging in developing an appropriate compost or fertilizer for soil amendment purposes [124]. Nonetheless, the authors claimed that their SCG composts, mainly without the addition of waste cardboard, can retain nitrogen due to the high oil content of SCG acting as an inhibitory factor on microbial degradation. In all the composts (after the composting process), the achieved C/N ratio was less than 25, suggesting proper nitrogen stabilization. The highest C/N ratio was for in-vessel composting (21.8), leading to the faster decomposition of organic matter [38].

Different SCG contents (0%, 10%, 20%, and 40% of dry matter) were studied to produce composts mixed with wheat straws and *Acacia dealbata*, which is an invasive plant in several parts of Europe. The authors reported a decrease in polyphenol and tannin compounds at the end of the process for all composts (both compounds decreased from 41.5 to 1.5 mg/g with 10% of SCG, from 38.5 to 0.6 mg/g with 20% of SCG, and from 32.5 to 0.9 mg/g with 40% of SCG). Soluble organic nitrogen values showed that higher coffee contents led to higher N contents from the start to the end of the experiment. The 40% SCG compost exhibited the highest total N content, which was 40% higher than the compost without

SCGs. The main novelty of this research was to assess the gas emissions of the composting process, which was associated with low emissions of CO_2, N_2O, and CH_4 during all processes. The authors concluded that 40% SCG provided the best conditions for the compost quality and gas emissions [13]. A further study investigated the toxicity and biological properties of biochar addition in SCG compost [8]. After 140 days, the biochar content resulted in a higher C/N ratio than SCG alone (12%), decreasing the biological formation (more than 100%) of bacteria and fungi, but allowed *E. coli* and *Salmonella* ssp. microorganism growth. As an important conclusion, biochar addition to SCG compost decreased the compost toxicity.

Two studies evaluated the effect of SCGs in composting. In the first study, they produced black compost via SCGs mixed with cow dung and chicken manure under aerobic conditions of fungi, *Bacillus*, and lactic acid bacteria. The results produced a black compost with a C/N ratio lower than the control sample (≤10) and pH values between 6 and 9. It was identified that *Bacillus* provided a better-decomposed compost but provided a low, stable compost (C/N ratio of 8.5:1) [37]. The second study investigated the reduction in SCG composting time through aerobic static batch composting (fungi and commercial activators) with temperature control to increase microorganism growth. After 28 days, FTIR spectroscopy showed a mature and nutrient-rich compost for fungi culture which was applied for radish production. The latter presented a germination index increases of only 4.2% compared with the commercial activator, but was 51.8% higher compared with the control sample. The authors concluded that aerobic batch composting could accelerate composting (shorter time) and maximize the process efficiency [19].

Furthermore, SCGs and coffee silverskin were applied in vermicomposting with horse manure. After 60 days of composting, it was observed that the waste content had an important effect on the results. Therefore, composts with 25% SCG and 25% or 50% coffee silverskin were found to be the best conditions with non-toxic behavior concerning biomass content, growth ratio, and germination [125]. It was emphasized that SCGs should be used with other amendments to increase the pH value [125], while avoiding toxicity from SCG components [7]. A recent study also investigated SCGs in vermicomposting aiming to develop a non-toxic compost. Straw pellets + SCG improved the growth of earthworms and increased the nutrient (P, K, and Mg) contents. Moreover, when vermicomposting was added to SCGs (25% and 50%) and straw pellets (75% and 50%), the caffeine decreased compared with SCG/straw pellets composting without earthworms (around 80%) [126].

A more recent study mixed SCGs with chicken and duck eggshells (200:20 g/g) and concluded that an SCG with chicken eggshell mix was the best in terms of improving the Ca, Mg, and K contents, as well as providing an adequate pH (5.96) [18]. Furthermore, SCGs were mixed with lightweight clay ceramic for an agricultural field with draining purposes (with 15 wt.% of SCG) [127] and as fertilizers for nursery grapevine production, providing water retention in the soil and improving leaf photosynthesis [128].

Therefore, either directly adding SCG to the soil or as an additive improving composting are two promising ways to add value to a mass-produced waste. Nonetheless, composting and vermicomposting are tested non-toxic ways of using SCGs in the soil for different agricultural applications.

5. Conclusions

SCG's main nutrients are K, P, Mg, and Ca, which play important roles in improving soil nutrients for agriculture. In fact, the advantages of using SCGs in agriculture are related to their nutrient range, which can fertilize the soil, accelerate plant growth, and even upgrade the nutrient content of the vegetables. In addition, SCGs used in agriculture prevent the improper disposal of this waste, decreasing the pollution caused by the degradation of the SCG's toxic components. However, the limitations for SCG uses are associated with its toxicity depending on the amount used, but this can be solved by composting mixtures with other wastes and apply some treatments to remove the toxic elements, such as caffeine and tannins. When used alone directly in the soil, the content should be around 10 wt.%.

Over the last 15 years, the literature has reported advantages and limitations of SCGs in agriculture. The toxicity of caffeine is well-established in the literature, but how to reverse this effect could be further explored. Moreover, the acceleration/inhibition of the plants' growth, types of soil and composting, and incubation time should be investigated because they are key to determining the optimal parameters for SCG applications. SCG biochar can also be a promising solution, as is combining SCGs with other coffee wastes and tea wastes. It was reported that SCGs combined with tea wastes could eliminate Fe deficiencies in soil. Finally, reduced gas emissions were reported, but new efforts should be made to quantify this effect, justifying the use of organic wastes to reduce environmental issues.

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