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# Heterogeneity in the Chemical Composition of Biofertilizers, Potential Agronomic Use, and Heavy Metal Contents of Different Agro-Industrial Wastes

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**Abstract:** Several agro-industrial, livestock, and food wastes can be recycled to create biofertilizers. This diversity of raw materials can result in nutritional imbalance and an increase in heavy metal content, which could make the final product unfeasible. Thus, the chemical characterization of the raw materials and their influence on the sustainable and safe production of biofertilizers need to be better understood. In this context, the objective of the present study was to evaluate the chemical characteristics of agro-industrial residues used in the manufacture of an aerobic liquid biofertilizer. We analyzed the macronutrient, micronutrient, and trace metal contents of seven waste products used as raw materials to create a biofertilizer. In addition, a survey of secondary biofertilizer data from different residues was carried out that showed great heterogeneity in the chemical compositions of these residues, which has a direct impact on the agronomic efficiency of these biofertilizers. The characterization revealed that some materials may be contaminants of the soil, due to high levels of trace metals, especially cadmium. We conclude that the generation of detailed inventories, such as those of the nutrient and heavy metal contents of the raw materials and biofertilizers produced, is indispensable for the correct recommendation of biologically-based inputs in agriculture.

**Keywords:** biologically based fertilizer; recycling; agroindustrial residues; trace metals

## 1. Introduction

The growing effort to intensify sustainable agricultural systems has resulted in increased use of biofertilizers [1,2]. A global assessment revealed that in 2016 alone, the biofertilizers market was approximately \$787 million [3]. This has resulted in a considerable reduction in the use of chemical fertilizers [4] and expanded the adoption of biologically-based inputs in agriculture.

It is already well-established that the use of biofertilizers can increase crop productivity. Although biofertilizers may vary considerably in response to the formulation used, the productivity of cereals, vegetables, legumes, and root crops may increase by 20% [5]. The inoculation of beneficial microorganisms, such as phosphate solubilizers increases biofertilizer potential and can increase crop production by up to 50% [6]. In addition to increasing crop yields, the use of biofertilizers can raise the levels of organic carbon and nutrients, as well as enrich the soil microbiota over time when compared with the use of conventional fertilizers [7].

Different biomasses, including residues from agriculture [8], industry [9], cattle raising [10,11], agroindustry [12,13], and domestic use [14], have been used in the preparation of biofertilizers. The diversity of raw materials is not only an environmentally sustainable solution to the management of different residues, but it also represents an excellent opportunity to recycle nutrients that can enrich the soils [15], especially those of tropical regions that present low fertility.

It is estimated that Brazilian agricultural activities, in the year 2017, used about 24 million tons of fertilizer. Despite the importance of agriculture for the Brazilian economy, more than 80% of the fertilizer used in 2017 was imported [16]. Therefore, the production of organic fertilizers from different residues is clearly a potential option to increase the economic, social, and environmental sustainability of Brazilian agriculture. With regards to agricultural residues, for example, Brazil ranks among the world's largest producers of cotton [17], castor oil, and corn [18], and is one of the major exporters of meat [19]. All of these activities generate large amounts of waste that represent excellent raw materials for the production of organic fertilizers. Despite these residues' potential for reuse and the large area devoted to agriculture (around 64 million ha, 7.6% of the national territory), Brazil is not even among the top 10 countries with regards to organic area [20], demonstrating the great growth potential of this activity and use of necessary inputs.

One of the hypotheses to explain the low use of organic inputs in agriculture is the heterogeneous chemical composition of the various available fertilizer formulations. It is possible that this lack of uniformity is related to the low availability of data regarding the chemical composition of these inputs and the raw materials, which makes it difficult to plan for the elaboration of formulations. In addition, due to different possible combinations among raw materials to obtain a biofertilizer, it is expected that there would be products with a wide range of nutrient contents [11,21–39]. In general, two major groups of waste sources have been used to produce biofertilizers: One is based on manure from different animals [21,22], and the other is composed of agro-industrial and household wastewater, such as cassava, olive, or domestic waste processing [23–39]. Even within the same group, chemical variability is still observed. For example, two types of bovine manure biofertilizers present a great variability of N concentrations, with values ranging from 1.8 to 8 g L<sup>-1</sup> [21,24].

Recent research has shown that the main reason for the second group of biofertilizers is the great diversity of available microorganisms (among which has been identified bacteria of the genera *Pseudomonas*, *Bacillus*, *Gluconacetobacter*, and *Corynebacterium*, as well as many other fungi and yeasts) [25]. When applied to plants, these biofertilizers stimulate physiological or natural processes that increase nutrient absorption, since such bacteria stimulate the production of metabolites, related directly to the growth of the plant (e.g., auxins, gibberellins, and cytokinins) [26]. They also synthesize antibiotics, siderophores, and hydrocinnamic acid that reduce pathogen activities [27]. In addition, these microorganisms further stimulate root development. *Pseudomonas* may also act as a phosphate solubilizer, increasing the soil-available P to plants [7]. As highlighted previously, the proposed biofertilizer provides not only a better balance among nutrient contents but also bio-stimulant properties. This can be seen as an advantage when compared to traditional biofertilizers with animal manures as a main ingredient.

Another important point is that the evaluation of contaminants, such as heavy metals in these inputs, which when present in high concentrations, may result in bioaccumulation in cultivated plants and consequently endanger the health of the final consumer [28]. Therefore, the chemical characterization of agro-industrial and domestic residues is of paramount importance in the preparation of more homogenous and safe formulations. This is in order to increase the reliable use of organic inputs, thus guaranteeing productivity indices and products that meet market requirements.

Given the scarcity of information regarding the chemical composition of different agro-industrial residues, the present work has the following objectives: (i) to chemically characterize agro-industrial residues used in the production of an aerobic biofertilizer and to evaluate its agronomic potential along with the heavy metal contents associated with such wastes; (ii) from this characterization

and the collection of secondary data, to systematize information on the nutrient contents of different biofertilizers used in agriculture in order to confirm the existence of heterogeneity in these formulations.

## 2. Materials and Methods

### 2.1. Chemical Characterization of the Agroindustrial Residues Used in the Aerobic Biofertilizer

The following agro-industrial wastes were evaluated: Cottonseed meal, castor bean meal, crushed legume seeds (soybeans, beans), corn meal, bone meal, blood meal, and wood ash. Samples of each material were ground and screened in a 0.074 mm mesh sieve for analysis of the biofertilizer raw materials. Three different samples of each of the aforementioned residues were evaluated in order to determine the measures of dispersion of the obtained data.

For the determination of macronutrients, micronutrients, and heavy metals, 500 mg of each material was subjected to nitric–perchloric digestion. After digestion, the samples were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES). The total N content was obtained by digestion with sulfuric acid and hydrogen peroxide, followed by subsequent distillation according to the Kjeldahl method [29]. Total organic carbon (TOC) was determined by combustion in a Perkin Elmer 2400 CHNS elemental analyzer.

### 2.2. Preparation of the Aerobic Biofertilizer

A formulation of the aerobic biofertilizer proposed by Embrapa Vegetables, called Hortbio<sup>®</sup>, was also used for comparison with the biofertilizers arising in the literature. This formulation was proposed for the purpose of valorizing the reuse of agro-industrial residues commonly produced in Brazil, with the intent of using it in vegetable production. It is a non-commercial product, the mode of preparation of which is available on the Embrapa website [30]. In this case, a biofertilizer formulation was prepared as described below, and aliquots of this formulation were then used to determine the chemical properties previously mentioned according to the protocols described below. The preparation of the biofertilizer was carried out in the organic agriculture sector and chemical characterization analyses were carried out at the Laboratory of Soil and Plant Nutrition of Embrapa Vegetables.

To prepare the biofertilizer, the following byproducts/residues were used: Blood meal (1.1 kg), cottonseed meal (4.4 kg), castor meal (1.1 kg), bone meal (2.2 kg), crushed seeds (1.1 kg), wood ash (1.1 kg), rapadura (0.55 kg), and corn meal (0.55 kg), enriched with 1 L of the Efficient Microorganisms (EM) inoculant. The EM inoculant was obtained from soil under native vegetation (15°56′61.8″S, 48°08′42.7″W), using cooked rice as a substrate, left in the soil under cerrado vegetation in a plastic tray for 10 days, according to the recommendations of the Book of Effective Microorganisms [31]. The EM was characterized as a mixture of bacteria (*Bacillus*, *Gluconobacter*, *Enterobacter*, *Acinetobacter*, *Pseudomonas*, and *Corynebacterium*), yeasts (*Pichia kudriarzerovi* and *Meyerozyma guilliermondii*), and fungi (*Penicillium*, *Aspergillus*, and *Trichoderma*) [25]. To these materials, non-chlorinated water was added to a final volume of 100 L. The final mixture was stored in a cool, shaded place with aeration for 15 min every hour using an air compressor and a timer.

### 2.3. Determination of the Chemical Properties of the Aerobic Biofertilizer

After preparation of the biofertilizer, 150 g aliquots were collected on the 10th day after the start of Hortbio<sup>®</sup> preparation. The samples were then filtered, frozen, and lyophilized for 12 h. After lyophilization, the samples were weighed, milled, and sieved in a 200-mesh screen. The chemical compositions of each raw material were determined by Inductively Coupled Plasma Spectroscopy (Model ICPE-9000, Shimadzu, Kyoto, Japan), following total solubilization of 0.5000 g with HNO<sub>3</sub>:HCl (3:1, v/v) in a microwave oven (CEM corporation, model marsxpress, Charlotte, USA).

After every 20 samples, a standard multi-element solution (Fluka Multielement—10 mg L<sup>-1</sup>) was used as a reference to correct for instrumental instability and matrix effect. All analyses were carried out in triplicate. The quantities of accumulated nutrients were obtained by the ratio between

the content of each nutrient and the percentage of dry mass, which was 6.8%. The determination of N and C was performed by combustion in a Perkin Elmer 2400 CHNS elemental analyzer.

#### 2.4. Acquisition of Data on the Chemical Composition and Heavy Metal Concentrations of Different Biofertilizers

##### Acquisition of Secondary Data

In order to understand the heterogeneity of biofertilizer formulations regarding nutrient supply and heavy metal concentration, data records were assessed for the following chemical attributes of these inputs: Hydrogen ionic potential (pH); electrical conductivity (EC); concentrations of C, N, P, K, S, Ca, Mg, Fe, Na, Zn, Cd, and Pb; and the C/N ratio. Data were collected from 14 different biofertilizer formulations.

#### 2.5. Statistical Analyses

The obtained dataset was submitted for evaluation of its dispersion by determining the mean, standard deviation, standard error, and coefficient of variation for each measured chemical property.

### 3. Results

#### 3.1. Chemical Composition of Agroindustrial Residue

The chemical compositions of the different residues, that are used to prepare the biofertilizer Hortbio<sup>®</sup> are presented in Table 1. The contents of macronutrients in the biofertilizer are also presented in order to facilitate comparison among materials. One can observe that the residues of cottonseed meal, castor bean meal, and blood meal had high N levels, equal to 5.57%, 6.72%, and 12.34%, respectively. Due to the higher values of P (44.88 g kg<sup>-1</sup>), Mg (58.91 g kg<sup>-1</sup>), and Ca (141.98 g kg<sup>-1</sup>), bone meal stood out as an important source of these nutrients for the biofertilizer composition. Wood ash represented the main source of K (84.44 g kg<sup>-1</sup>) and Ca (175.32 g kg<sup>-1</sup>). However, depending on the source of this raw material, these values can change. Among all the raw materials used, corn meal had the lowest nutrient contents.

**Table 1.** Chemical properties of the different raw materials used in elaboration of the aerobic biofertilizer.

Materials <sup>a</sup>	N	C	P	K	Ca	Mg	S	Na
	%		g kg <sup>-1</sup>					
Cottonseed meal	5.57 ± 0.98	32.32 ± 0.98	7.45 ± 0.22	16.58 ± 0.67	2.06 ± 0.03	4.46 ± 0.18	2.78 ± 0.04	0.38 ± 0.05
Castor bean meal	6.72 ± 0.29	27.94 ± 0.25	7.50 ± 0.34	14.44 ± 0.61	6.40 ± 0.08	5.37 ± 0.18	4.03 ± 0.19	0.54 ± 0.10
Corn meal	1.11 ± 0.50	-	1.13 ± 0.06	3.80 ± 0.10	0.19 ± 0.03	0.52 ± 0.004	0.83 ± 0.01	0.49 ± 0.02
Blood meal	12.34 ± 0.49	35.37 ± 0.43	2.32 ± 0.38	5.40 ± 0.18	5.26 ± 0.42	0.22 ± 0.02	5.01 ± 0.25	15.42 ± 0.5
Seeds meal	4.88 ± 0.03	38.91 ± 1.57	4.35 ± 0.48	34.83 ± 0.79	4.02 ± 0.33	2.53 ± 0.34	2.62 ± 0.50	1.04 ± 0.01
Bone meal	0.03 ± 0.01	2.22 ± 0.01	44.88 ± 1.55	ND	141.98 ± 13.2	55.55 ± 4.30	4.34 ± 0.06	3.49 ± 0.18
Ash	0.040 ± 0.01	-	7.62 ± 0.07	84.44 ± 1.25	175.32 ± 3.42	34.53 ± 2.33	6.64 ± 0.18	21.62 ± 0.16
Biofertilizer	0.45	20.00	0.81	2.93	3.91	1.00	0.46	-

ND = not detected; mean values ± standard error; n = 3; <sup>a</sup>: agroindustrial residues.

##### 3.1.1. Micronutrients of Agro-Industrial Waste Properties

Agricultural production limitations, caused by micronutrient deficiency, have been reported in soils from different parts of the world, including South America [32] and India [33]. The sources characterized in the present study contained high levels of micronutrients such as Fe, Mn, and Zn (Table 2). Agro-industrial residues, such as blood meal and ash had high Fe contents, with values of 3453.33, and 15133.33 mg kg<sup>-1</sup>, respectively. Greater amounts of Mn (1096.67 mg kg<sup>-1</sup>) and Zn (276.77 mg kg<sup>-1</sup>) were also supplied by the ash. However, corn meal residue contained the lowest Fe, Mn, and Zn contents among all the characterized residues.

**Table 2.** Micronutrients in different raw materials used in elaboration of the Hortbio® biofertilizer.

Materials <sup>a</sup>	Fe	Mn	Co	Cu	Zn
	mg kg <sup>-1</sup>				
Cottonseed meal	80.80 ± 6.51	14.30 ± 0.09	ND	ND	50.91 ± 1.56
Castor bean meal	202.67 ± 20.66	54.93 ± 2.28	ND	ND	109.44 ± 3.81
Corn meal	63.13 ± 2.08	2.65 ± 0.06	ND	ND	11.81 ± 0.61
Blood meal	3453.33 ± 124.93	13.67 ± 0.94	ND	ND	27.77 ± 1.49
Crushed seeds	172.67 ± 29.01	29.97 ± 5.00	ND	ND	58.44 ± 6.43
Bone meal	823.67 ± 117.3	27.33 ± 0.97	ND	ND	ND
Ash	15133.3 ± 777.9	1096.67 ± 2.72	ND	706.40 ± 85.96	276.77 ± 21.56
Biofertilizer	121.15	ND	ND	ND	8.76

ND = not detected; mean values ± standard error; *n* = 3; <sup>a</sup>: agroindustrial residues.

### 3.1.2. Heavy Metals Analysis of Agro-Industrial Waste Properties

Table 3 shows the heavy metal contents of the different organic sources, and the maximum values allowed by the Ministry of Agriculture, Livestock, and Food Supply of Brazil (MAPA), Decree No. 4.954 of 2004, which regulates Law No. 4.954 of 1980 [34] and dictates inspection of the production and trade of fertilizers, correctives, inoculants, or biofertilizers intended for agricultural use. In addition, the heavy metal contents of the biofertilizer produced from the raw materials are also presented in Table 3. It was observed that of all the residues characterized, castor bean meal and crushed seeds had high levels of Cd, equal to 109.9, and 26.7 mg kg<sup>-1</sup>, respectively. In the cottonseed and bone meal residues and the ash, the contents of Cd were below the limit of quantification (LOQ) of the analytical method.

**Table 3.** Heavy metals in different raw materials used in the elaboration of biofertilizers.

Heavy Metals	Cottonseed Meal <sup>c</sup>	Castor Bean Meal <sup>c</sup>	Corn Meal <sup>c</sup>	Blood Meal <sup>c</sup>	Seeds Meal <sup>c</sup>	Bone Meal <sup>c</sup>	Ash	Decree 4.954 <sup>a</sup>	CONAMA No. 460 <sup>b</sup>	Hortbio <sup>d</sup>
mg kg <sup>-1</sup>										
Cd *	LOQ	109.9 ± 0.0	0.91 ± 0.36	4.9 ± 0.6	26.7 ± 2.3	LOQ	LOQ	3	3	0.1
Cu	LOQ	LOQ	LOQ	LOQ	LOQ	LOQ	706.4 ± 85.9	-	200	LOQ
Ni	LQ	LOQ	LOQ	LOQ	LOQ	LOQ	LOQ	70	70	LOQ
Pb	LOQ	LOQ	3.9 ± 1.3	LOQ	LOQ	LOQ	LOQ	150	180	LOQ

LOQ = below the limit of quantification; mean values ± standard error; *n* = 3; <sup>a</sup>: Decree no. 4.954 of 2004: Maximum limits of contaminants allowed in organic fertilizers and soil conditioners; <sup>b</sup>: The National Environment Council—CONAMA, Resolution 420 of 2009: Maximum values of contaminants allowed in the soil. <sup>c</sup>: agroindustrial residues; <sup>d</sup>: biofertilizer. \* Limit of quantification (LOQ): Cd > 0.03 mg L<sup>-1</sup>, Ni > 0.05 mg L<sup>-1</sup>, and Pb > 0.04 mg L<sup>-1</sup>.

### 3.2. Physical and Chemical Characteristics of Biofertilizers Obtained from Different Raw Materials

This approach facilitated the comparison between different residues of vegetal, animal, agro-industrial, or urban origins. Table 4 presents a compilation of the chemical compositions of several biofertilizers produced from different formulations.

**Table 4.** Chemical characterization of biofertilizers obtained from different agroindustrial waste sources.

Biofertilizer	pH	EC	C	N	C/N	P	K	S	Ca	Mg	Fe	Na	Zn	Cd	Pb	Source
	dS m <sup>-1</sup>			g L <sup>-1</sup>						mg L <sup>-1</sup>						
1	6	5.9	200.3	4.5	4.5	0.81	2.93	0.46	3.91	1	121.15	-	8.76	0.1	-	Biofertilizer in study
2	7.6	11.2	-	1.8	-	0.43	2.7	-	-	-	-	-	-	-	-	[21]
3	6.27	-	-	87	-	6.3	14.7	5.5	0.31	0.52	-	9600	1.58	-	-	[35]
4	8.02	-	-	2.2	-	0.23	1.13	0.1	0.73	0.14	-	640	1.15	-	-	[35]
5	-	-	1.4	0.2	7	-	0.075	-	0.55	0.13	0.01	-	-	-	-	[22]
6	6.8	-	15.4	1.64	9.39	0.068	0.42	-	-	0.064	6.5	-	1.6	0.23	0.87	[36]
7	5.25	7.1	-	8	-	0.4	6.95	-	1.08	0.72	-	-	-	-	-	[24]
8	-	6.16	-	-	-	-	0.82	0.19	0.45	0.12	-	205.67	-	-	-	[37]
9	-	-	-	1.8	-	0.25	3	0.35	-	0.12	-	350	-	-	-	[23]
10	6.77	5.34	4	0.5	8	0.54	0.32	-	0.217	0.12	9.56	130	-	-	-	[26]
11	-	-	-	1.4	-	0.8	0.42	0.6	4.93	0.55	1.02	-	5.33	0.13	0.33	[38]
12	-	-	-	2.2	-	0.93	0.3	0.5	2.8	0.42	0.62	-	3	0.07	0.22	[38]
13	5.01	6.93	30.5	20.7	2	2.22	19.24	-	-	-	-	33980	-	-	-	[39]
14	7.2	9.69	18	2.1	8.62	0.002	1.4	-	3.75	0.003	0.18	-	62	-	-	[40]
15	8.1	-	2.3	0.4	6	0.5	0.7	0.24	0.58	0.26	6.36	-	3.72	-	-	[11]
CV (%)	14	32	160	222	34	150	152	162	98	90	203	170	168	41	52	

1 = Aerobic biofertilizer of the present study; 2 = Sheep manure (50%) and cattle manure (50%); 3 = Hydrolyzed salmon protein; 4 = Anaerobic digestion of urban residues; 5 = Pig manure; 6 = Sweet potato starch wastewater; 7 = Fresh dairy cow manure, mineral phosphate, legume seed meal, wood ash, milk, sugar; 8 = Manure of dairy cows resting for 30 days, sugarcane molasses, milk, agricultural gypsum; 9 = UG max; 10 = Rabbit manure; 11 = Aerobic sludge resting for one year (autoclaved); 12 = Fresh aerobic sludge (autoclaved); 13 = Cassava peel and wastewater from cassava processing; 14 = Olive wastewater and dehydrated poultry manure; 15 = Breeding pig wastewater.

## 4. Discussion

### 4.1. Chemical Properties of Agroindustrial and Heavy Metal Concentrations of Raw Materials

The N content in the residues of blood meal, castor bean meal, and crushed seeds exceeded the mean levels present in fresh cattle, buffalo, and goat manures by 20, 11, and 8 times, respectively, when characterized by Nath et al. [41]. Even with high levels of N after the 35-day resting period, cattle manure presented N content that was ten times lower than this nutrient in the blood meal residue of the present study [24]. The characterization of urban food residues showed N contents similar to those observed in corn meal residue and five times lower than the N content of blood meal residue [42]. When compared with seafood residues, the blood meal also presented twice as much N in its composition [43]. With the characterization of other nutrient sources, it is possible to substitute a lower concentrated material for another more enriched material, facilitating fertilization practices by reducing the applied amounts, as observed in the work of Chatterjee [44]. In this study, for a lettuce yield of 11 t ha<sup>-1</sup>, approximately 20 t ha<sup>-1</sup> of bovine manure was applied. Despite the difference in cost of each input, the use of bone meal residue would reduce the dose by 20 times (i.e., it would be necessary to apply only 1.0 t ha<sup>-1</sup>). It is also important to study other sources of the nutrient in question because there are no nitrogen-rich mineral sources that are permitted by Brazilian legislation in organic agriculture, unlike other nutrients such as P, K, Ca, and Mg, which are found in mineral sources, such as phosphate, potassium, and limestone rocks.

The total P content of the bone meal residue found in the study was 45 g kg<sup>-1</sup>, lower than the 50–160 g kg<sup>-1</sup> range presented in the work of Möller [45]. According to Mattar et al. [46], bone meal residue is a source that can potentially be used as a fertilizer and consists predominantly of hydroxyapatite (Ca<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>(OH)). Phosphorus is the second most important nutrient in agricultural production, and because it is a non-renewable and non-replaceable mineral, it demands strategies that allow for reuse or recycling of this nutrient [47]. Other forms of transformation, such as ash or biochar of bones, enrich the contents of this material, surpassing the values found in the present study by two times [45,48]. Ylivainio et al. [49] tested bone meal residue, cattle manure, and superphosphate as sources of P in pasture cultivation and showed similar production after the third year of application. Similarly, Chen et al. [46,50] used conventional fertilizer and meat-and-bone meal residues in the cultivation of wheat and barley without showing significant differences in production of the two crops.

The main source of K was the wood ash among the studied materials (84.44 g kg<sup>-1</sup>). In addition to ash, the seed meal residue represented the second richest source of K (34 g kg<sup>-1</sup>). The K content of ash in the present study was two to three times higher than that found in spruce ash and the mean ash value of a large database in Sweden [51,52]. Basak et al. [53] and Lima et al. [54] characterized mica feldspar and biotite rock powders, respectively, and obtained K contents close to 16 g kg<sup>-1</sup>, which, when compared with the contents presented in this study for wood ash and crushed seed residue, are six, and two times lower, respectively. When compared with soluble mineral sources such as KCl, the K contents in wood ash and crushed seed residue are about five times smaller. On the other hand, composted chicken bed obtained by Mekki et al. [39] presented lower levels of K compared with the main sources of this nutrient in the present study (ash and seed meal residue). In agriculture, potassium rocks are the main source of potassium used in the manufacture of fertilizers, but their environmental impacts, including the use and emissions of greenhouse gases during mining, are of concern [55]. In 2010, Brazil imported approximately 90% of this mineral [56]. Initiatives to partially replace this nutrient with other raw materials considered residues from different activities can minimize these impacts.

The main sources of Ca in the present study were bone meal residue and ash, with levels of 14%, and 17%, respectively. Vestergård et al. [52] and Qin et al. [57] characterized ash from wood residues and found Ca contents close to those obtained in the present study of 12%. Mattar et al. [46] reported higher Ca concentrations (30%) in bone ash. Kolahchi and Jalali [58] characterized eight different agro-industrial residues and found Ca contents of 0.4% in beet residues and 8% in avian manure.

Wood ash is typically applied to soils to neutralize acidification and depletion of promoted cations by the addition of exchangeable bases present in the material [59]. Consistent research on the benefits of adding ash from different residues (wood, sugarcane bagasse, rice husk, and olive) reveals the positive influence of its addition on crops. For example, in the soil, there has been found to be an increase in pH [53,57] and organic carbon and its stocks [60]. In the case of inorganic carbon, there was also a decrease in CO<sub>2</sub> emissions [61], an increase in macronutrients [59], and a consequent higher crop productivity.

The highest levels of Na were obtained in the ash, followed by blood meal, bone meal, and crushed seed residues. The other raw materials presented Na values lower than 1 g kg<sup>-1</sup>. The presence of Na in plants aids in cell expansion, water balance, and osmotic regulation [62]. Despite its importance, the elevated presence of this element contributes to an increased biofertilizer salinity, thereby increasing the electrical conductivity of the soil, and significantly reducing the yield of most crops. Vegetables are generally sensitive to increased soil electrical conductivity [63]. Salinity is a complex phenomenon that affects the metabolic processes of plants, altering physiological and biochemical parameters [64].

In the present study, the main sources of Zn were ash (200 mg kg<sup>-1</sup>) and castor bean meal residue (100 mg kg<sup>-1</sup>). Studies performed by other authors indicated additional sources of Zn: Pig and chicken manure, with levels varying from 65 to 72 mg kg<sup>-1</sup> [65]; cattle manure, with levels close to 250 mg kg<sup>-1</sup> [66]; and urban waste, with a level of 30 mg kg<sup>-1</sup> [42].

In England in 2002, approximately 40% of Zn inputs for agricultural production were supplied by manure from different livestock activities [67].

In the materials considered, the main sources of Fe—the ash and blood meal residue—presented contents of 15 and 3 g kg<sup>-1</sup>, respectively. Mondal et al. [68] found Fe levels in urban waste vermicompost and cattle manure of 0.9, and 0.7 g kg<sup>-1</sup>, respectively. The Fe content in commercial products that supply micronutrients, such as Quelatec, is generally 7.5% Fe. This is five times higher than the concentration found in wood ash, the main source of iron used in the present study.

These high Cd levels in castor bean meal residue are alarming because this chemical element is highly toxic and widely used in the formulation of biological inputs. Castor bean (*Ricinus communis*) is recommended as a crop for the remediation of areas containing Cd, due to its bioaccumulation [69]. Baudhdh and Singh [70] also reported high levels of Cd prevailing in castor bean meal, indicating its potential as a good phytoremediator or hyperaccumulator for several toxic organic and inorganic chemicals.

The presence of trace metals above the permitted limits may limit the use of the different characterized raw materials. The use of some residues with high metal contents can lead to soil contamination, in addition to the potential accumulation of these metals in the roots of the plants [71]. In this sense, castor bean meal residue presents a risk of contamination due to the high levels of cadmium. Considering lettuce cultivation as an example, 150 kg ha<sup>-1</sup> of N is required for a yield of 2.1 t ha<sup>-1</sup> [72]. If all this N were applied using castor bean, 279.8 mg of Cd would be added to the soil, exceeding in a single application the maximum limit of 3 mg kg<sup>-1</sup> of soil, as recommended in The National Environment Council (CONAMA) Resolution no. 460 [73].

#### 4.2. Acquisition of Data on the Chemical Composition Different Biofertilizers

The variation observed in the composition of the different biofertilizers clearly showed a direct relationship with the raw materials used. Using the coefficient of variation as a reference, the highest variations encountered were with respect to the concentrations of N (222%), followed by Fe (203%), Na (170%), S (162%), C (160%), Zn (168%), K (152%), P (150%), Ca (98%), Mg (90%), Pb (52%), and Cd (41%); C/N ratio (34%); electrical conductivity (32%); and pH (14%). These results show the chemical composition variability of the biofertilizers reported in scientific literature, which in turn implies the need for adequate fertilization management as a function of the input produced.

The variability of the C/N ratio was also relatively high, indicating that the biofertilizer decomposition rate, with respect to the nutrient release of agricultural crops should also be different.

This information is important because it can define more promising inputs for use in short- or long-cycle crops. Thus, inputs that present a lower C/N ratio should release nutrients faster, making them more suitable for short-cycle crops such as vegetables. On the other hand, those with higher C/N ratios can be used for long-cycle crops because the nutrients are gradually released. Comparing the chemical characteristics of the biofertilizers indicates there is a great variability because 60% of the time they were used in cereals, 20% in vegetables, and 20% in fruits.

Among the biofertilizers detailed in Table 4, two major groups of waste sources were observed: The first based on the manure of different animals, and the second was composed of agro-industrial and household wastewater, such as cassava, olive, or domestic waste processing. The mean concentrations of the major nutrients N, P, K, and Ca for the first group were 3.8, 0.5, 1.7, and 1.2 g L<sup>-1</sup>, respectively. In the second group, the concentrations were generally higher for all of the nutrients (5.72, 4.1, 5.05, and 3.05 g L<sup>-1</sup>), and the biofertilizer consisting of cassava residues was one of the largest suppliers of N and P. The studied aerobic biofertilizer had the fourth highest potential to provide N with a concentration of 4.5 g L<sup>-1</sup>, following the biofertilizer consisting of hydrolyzed salmon protein (87 g L<sup>-1</sup>), the biofertilizer of cassava process residues (20.7 g L<sup>-1</sup>), and the biofertilizer of bovine manure (8 g L<sup>-1</sup>).

When observing the concentrations of two bovine manure biofertilizers (numbers 2 and 7), there was great variability of N concentrations among these materials, with values of 1.8 and 8 g L<sup>-1</sup>, respectively. In general, heterogeneity in the composition of biofertilizers is linked to the chemical and physical characteristics of the raw material, such as the initial C/N ratio of these materials [74], EC, and pH values [75]. Kiehl [76] affirmed that organic fertilizers can vary greatly in composition, rate of mineralization, and available N content, factors that in turn are influenced by environmental conditions and requiring more attention in characterization.

Another noticeable point was the lack of information on the presence or absence of heavy metals in the different biofertilizers. In England, an inventory of major sources, with possible concentrations of trace metals, such as sewage sludge, manure, lime, agrochemicals, agro-industrial waste, and composts, showed that the continued use of wastes, such as sewage sludge and manure would increase the concentrations of heavy metals to the maximum allowable limit in a shorter time [66]. The variability of sources may explain the decline or instability of production, with a direct consequence on the profitability of the producer. In addition, the lack of testing and information in the search for standardization limits its exploitation. Therefore, the search for mechanisms to standardize the formulations becomes essential. Finally, an alternative would be to work with recommendations using the lowest value observed to ensure adequate supply of a particular nutrient.

## 5. Conclusions

Despite the Brazilian agribusiness being responsible for almost 30% of the GDP, its vulnerability is a common economic concern, since Brazil imports almost all NPK fertilizer. That scenario is more sensitive for sustainable production systems (i.e., organic or agroecological farming), where the number of fertilizer options are limited. Because of that, searching for other sustainable alternatives, such as biofertilizers produced by recycling agro-industrial byproducts, may contribute to reducing external dependence on fertilizers. Due to the great variability of sources and nutrient contents of the raw materials, the chemical composition of such biofertilizers is uncertain, and the presence of heavy metals is an environmental concern.

Our results confirm that there is a lack of standardization in the nutrient content of different biofertilizers as a result of different formulations. By comparing the raw materials of our fertilizer with others reported in the literature, it was possible to separate them into two major groups: One based on manure from different animals and another composed of agro-industrial or household wastewater, the latter having higher concentrations of the major nutrients N, P, K, and Ca than the former. The high Cd levels in castor bean meal residue are alarming because this element is highly toxic and this residue is widely used in the formulation of biological inputs. It was also concluded that the generation of

detailed inventories with the chemical compositions of the raw materials and produced biofertilizers is a prerequisite for the correct recommendation of biologically based inputs in agriculture.

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