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Levels of selected metals in spices cultivated in Southwest Ethiopia: Occurrence and health risk assessment

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ABSTRACT

In this study, the levels of seven metals (Ca, Cd, Cr, Cu, Zn, Pb, and Ni) in the soil and spice plants grown around Oromia Region of Southwest Ethiopia were analyzed using flame atomic absorption spectrometry (FAAS) after acid digestion. The pH of the soil samples was found to be in the range of 5.86 to 6.57, with organic matter 13.93 to 20.59% and electrical conductivity 0.28 to 0.49 mS/m. Among the metals determined, Ca was the most prevalent, ranging from 429.01 to 5369.67 mg/kg, Zn 31.88 to 67.70 mg/kg and Cu 7.62 to 10.67 mg/kg in plants, while Cd, Cr, Pb, and Ni were not detected in the spice plants. Ca was the most abundant metal in soils, ranged from 1195.67 to 4147.17 mg/kg, followed by Zn, Cr, Ni, and Cu, ranging from 112.90 to 120.25 mg/kg, 21.92 to 45.76 mg/kg, 21.40 to 56.93 mg/kg and 13.03 to 28.67 mg/kg, respectively. Cd and Pb were not detected in the soil samples. The hazard quotient and hazard index values of Cu and Zn in all spices were below one, which may ensure that these spices do not pose substantial health risks to consumers.

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1. Introduction

Spices are plant parts that are used as food additives to improve the flavor, palatability, color, and acceptability of foods [1]. They are often used as food flavorings and sometimes as preservatives to prevent harmful bacterial growth [2]. Food contamination is the major route through which harmful metals enter the human body, making it imperative to monitor the bioavailability of metals in contaminated plants. Rapid industrialization, population growth, insufficient pollution control mechanisms, and improper utilization of chemical fertilizers and pest control mechanisms are the main factors responsible for heavy metal contamination in agricultural soils. As a result, toxic heavy metals from contaminated soils may accumulate in plants and enter the human food chain [3].

Plant contamination with heavy metals originated primarily from contaminated soils [4]. However, the environmental conditions of the cultivated farms and agricultural practices contribute significantly to the level of heavy metals that can accumulate in plants used as spices for food, functional foods and nutritional or nutritional supplements [5].

Essential metals are needed by plants in adequate quantities for their growth [6]. Most of the metals that enter the soil could be potentially toxic not only to plants but also to soil microbes and the soil ecosystem. Furthermore, recent studies have confirmed the increased research interest focusing on the accumulation levels of heavy metals in air, water, soil, and food due to their harmful effects on human health and aquatic lives [7-9].

In addition, prolonged intake of foodstuffs with high levels of heavy metals can result in chronic accumulation of these metals in the liver and kidneys of humans, leading to a variety of issues in numerous biochemical processes and ultimately leading to kidney, cardiovascular, nervous, and bone diseases [10,11]. The risks associated with the consumption of spices depend on the amounts of spices consumed and the average weight of the individual consumer. In most countries, consumers of spices are exposed to heavy metals through both consumption and skin contact, which can lead to adverse health effects.

In recent years, numerous efforts have been devoted to determining the metal content in spices and herbs [12]. Flame atomic absorption spectrometry (FAAS) is commonly used to the determination of metals in plants and soils, typically following wet digestion for sample preparation across various matrices [13-15]. In addition, there are quite a number of reports in the literature that trace level heavy metal contaminants are selectively separated from complex matrices

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employing different sample preparation methods, including metal chelate formation [16], cloud point extraction [17], dispersive liquid-liquid microextraction (DLLME) [18], and the use of low-cost adsorbents [19], although applications are limited to a single or a few metallic elements that are similar in certain physicochemical properties.

Ethiopia is among the countries in East Africa that produce and use spices heavily during food preparations, not only for flavoring purposes but also as traditional practices, which could also greatly help in food preservation for longer periods [20]. It is well known and documented that plant materials and therefore ready to use spices accumulate considerable amounts of toxic heavy metals in different parts of their tissues, which originated primarily from the soils [21]. As the level of the metals is not known precisely, consumers may be endangered by higher concentrations of the various metal constituents. In most cases, since the symptoms of their toxicities are usually not acute, the extent of the risks could therefore be worsened. To date, there are no reports in the literature showing the levels of metals in the spices grown in the districts of the Jimma Zone of the Southwest Ethiopia. This study was thus primarily aimed to determine the levels of selected heavy metals and assess the potential health risks associated with the consumption of widely cultivated spices in Southwest Ethiopia, including kororima (*Aframomum corrorima*), ginger (*Zingiber officinale*), turmeric (*Curcuma longa)* and black pepper (*Piper nigrum*).

2. Experimental

2.1. Chemicals and reagents

The study used high-quality analytical grade chemicals and reagents, including HNO_3 (69-72%), Sigma Aldrich (Streinheim, Germany), $HClO_4$ (70%) obtained from Research Laboratory Fine Chemical Industry (Mumbai, India), H_2O_2 (30%) provided by BDH Chemicals Ltd (Poole, England) and HCl (36-38%) purchased from Sigma Aldrich (Streinheim, Germany). These chemicals were used to digest spices and soil samples. Lanthanum nitrate hydrate, 99.9%, obtained from Hopkin and Williams Ltd (Essex, England) was utilized to avoid chemical interference during the determination of Ca. Buck Scientific Puro-Graphic (USA) provided the standard stock solutions of the target metals, with concentrations of 1000 mg/L. Double distilled and deionized water of chemical conductivity less than 1.5 µS/m was used for sample solution preparation, dilution and washing of the apparatus prior to analysis.

2.2. Apparatus and instruments

An air circulating oven, PLT-125, was used for sample drying, while a ceramic mortar and pestle (Hadenwanger, Germany) were utilized for grinding and homogenization of the samples. Digital electronic balance (PW-124, UK) was used to weigh the samples. A Kjeldahl apparatus (Gallenkamp, England) was used for the acid digestion of the spices and soil samples, while a pH meter, obtained from Adwa, model AD-1020, Romania, was used for pH adjustments and electrical conductivity measurement of the soil samples. Finally, the metal contents of the soil and plant samples were determined using a flame atomic absorption spectrophotometer (FAAS) (Model 700P Zeenit, Germany) with hollow cathode lamps and air-acetylene flame.

2.3. Study area

The spices and their corresponding plant parts, from the edible section, were obtained from Jimma Administrative Zone of Oromia Regional State, Southwest Ethiopia. The location is situated between 7^{0}40'42"N latitude and 36^{0}49'24"E longitude, with an altitude of 1,780 m above sea level. The selection of sampling sites was based on the quantities of spices produced compared to other spice producing regions.

2.4. Sample collection and preparation

The spice sample collection was carried out twice; the first in January 2014 and the second in January 2020. The second time sampling was planned, since the health risk assessment was necessary to be studied and evaluated on the freshly collected spice samples. The same metals were considered during both analyses. The spices; korarima, ginger, turmeric, and black pepper, were carefully selected from designated study areas and transported to the Analytical Research Laboratory of Addis Ababa University (AAU) in clean polyethylene bags. To remove any contaminants or airborne pollutants, the fresh spice plant samples were thoroughly washed with double distilled and deionized water and then air-dried for a week. The samples were oven-dried at 70-80 °C for 24 h to remove moisture. The dried samples were ground using a mortar and pestle and sieved using a 0.425 mm sieve size to prepare a fine powder of spices for the digestion step. The sieved samples were placed in polyethylene bags and then in desiccators at room temperature.

Soil samples were collected from the origin of the spice plants at a depth of 15 cm using a stainless-steel auger and the sample collection was carried out following the procedure used in the published work [22]. The soil samples were dried in the open air and then oven dried until the weight was constant. Subsequently, the samples were ground and sieved into a course and fine fractions; with the fine fractions were thoroughly mixed and homogenized using mortar and pestle and sieved (using a 0.425 mm sieve). Finally, the resulting homogenized samples were stored in a clean and dry container until digestion.

2.5. Digestion of the spice samples

For efficient breakdown of the plant materials, 0.5 g of each powdered, sifted, and homogenized sample was weighed and transferred to a 100 mL round bottom flask. Then, an acid mixture was prepared containing 4 mL of HNO_3, H_2O_2 and $HClO_4$ in a volume ratio of 2:1:1, according to the optimized procedure. The mixture was then added to the flask and digested on Kjeldahl digestion apparatus at 270 °C for 3 h, until a clear solution was obtained. After cooling, a 5 mL portion of the double distilled and deionized water was added to each sample in order to dissolve the precipitates that occurred during dissolution, and the solution was gently swirled. The resulting mixture was filtered through a Whatman Number 42 filter paper to remove any suspended or turbid materials, and the filter paper was then rinsed with double distilled and deionized water to dilute the sample to a final volume of 50 mL. This process was repeated three times for each plant sample. The digested, filtered, and diluted sample solutions were finally analyzed by FAAS.

2.6. Digestion of the soil samples

The soil samples collected from the locations where the spices were collected have undergone acid digestion. This was achieved by treating a 0.5 g dried, homogenized, and powdered soil sample into a 100 mL conical flask, followed by the addition of a 5 mL acid mixture comprised of HNO_3, HCl, and $HClO_4$ in a 3:1:1 volume ratio. The concoction was then digested using a Kjeldahl digestion apparatus at a temperature of 240 °C for a period of 2 h, until a transparent solution was produced. Three separate digestions were performed for each sample. The extracted solutions were filtered and diluted in a 50 mL volumetric flask with deionized water and refrigerated until further analysis [23].

2.7. Determination of heavy metals

The concentrations of Ca, Cd, Cr, Cu, Pb, Ni and Zn in the filtrate of the digested spices and soil samples were determined utilizing FAAS, which was equipped with deuterium arc background connectors and hollow cathode lamps and air acetylene flame. The FAAS was calibrated using the four working standard solutions. The working standard solutions of all metals were freshly prepared from intermediate standard solutions (100 mg/L) that were diluted from stock solutions of 1000 mg/L of the analyte metals. A deuterium background correction was made for all metals considered in the study.

2.8. Method detection limit

The minimum amount of analyte that can be determined with a specific level of confidence is called the detection limit. To determine the detection limit, eight blank samples were run to determine the mean and standard deviation of the blank. For spice and soil samples, eight replicate blank samples were analyzed for their respective heavy metal contents. The mean and standard deviations of the blanks were calculated to determine the method detection limit (MDL) which was calculated based on Equation 1 [24].

$$MDL = Y_B + 3SD_B \tag{1}$$

where Y_B is the analyte concentration in the blank solution and SD_B is the standard deviation of the replicate measurements of the analytes in the blank.

2.9. Evaluation of the analytical method performance

The spices and soil samples were spiked with a known amount of standard metal solutions independently in triplicate. The resulting spiked samples were then analyzed for their respective metal contents using FAAS. To calculate the percent recoveries for both the plants and soil samples, Equation 2 was used:

$$\mathrm{R}_A = \frac{Q_A(O+S) - Q_A(O)}{Q_A(S)} \tag{2}$$

where Q_A(S) is the spiked quantity of analyte 'A', Q_A (O+S) is the recovered quantity of analyte A from the spiked sample, and Q_A(O) is the quantity of the analyte in the original sample [25].

2.10. Determination of the soil pH

The pH of the soil was measured using a pH meter, with a suspension ratio of 1:2.5 (w/v) between the soil and the double distilled and deionized water. Five grams of air-dried soil, with a particle size of less than 0.425 mm, were weighed and placed in a 100 mL beaker. Then, 12.5 mL double distilled and deionized water was added and swirled for 5 min. Finally, the measurement was carried out after allowing the suspension to stand for 10 min at room temperature [26]. The process was repeated three times and the mean values were recorded.

2.11. Determination of soil organic matter

The organic matter (OM) of the soil sample was determined using a stainless-steel crucible rack and an electronic balance with accuracy of 0.0001 g. To obtain a constant weight, the sample was dried at 105 °C before determination of the OM content. The soil sample was weighed and heated at 450 °C for 4 h in the furnace, and then cooled in a desiccator and reweighed. The percentage OM was calculated using Equation 3 [27].

$$\mathrm{OM}\ \% = \frac{Wt\ (105^{\circ}C) - Wt\ (400^{\circ}C)}{Wt\ (105^{\circ}C)} \times 100 \tag{3}$$

where OM is the organic matter; W_t is the weight of the soil samples at a specified temperature. This measurement was carried out in triplicate and the mean values were reported.

2.12. Determination of electrical conductivity of soil

The measurement of electrical conductivity (EC) of the soil sample was carried out using a conductometer, with a 1:5 (v/v) mixture of soil and double distilled and deionized water. First, 5 g of the soil sample was weighed and transferred to a beaker. Then, 25 mL of double distilled and deionized water was added to the beaker and a magnetic stirrer was used to create a uniform and homogeneous soil sample solution. Electrical conductivity was measured after standing for 10 min at room temperature [28]. It is important to note that all measurements were performed in triplicate and the mean values were reported.

2.13. Health risk assessment of spices consumption

The consumption of spices may pose health risks due to the presence of heavy metals and it is important to assess these risks. These risks are typically determined based on the estimated daily intake (EDI) of the metal, the target hazard quotient (THQ), and the hazard index (HI). The EDI values of the analyzed metals were calculated using Equation 4 [29].

$$EDI = C^{*}{}_{metal}\,\frac{IR}{BW} \tag{4}$$

where EDI is the estimated daily intake and C_{metal}, in mg/kg, is the average weight of the heavy metal contents of the spices, IR (ingestion rate) is the average daily consumption of spices in grams/day/person, and BW is the average body weight of the consumer, in kilograms.

The average IR for spices for adults is 10 g/day/person of dry weight, and the average body weight for adults taken in this study was 65 kg [30-32]. The THQ is used to assess non-carcinogenic risks associated with long-term exposure to contaminants in spices, and the THQ evaluation was made using Equation 5:

$$THQ = \frac{EDI}{RfD} \tag{5}$$

where THQ is the target hazard quotient and RfD is the reference dose for the metal, in mg/day; RfD values for Cu and Zn are 0.04 and 0.30 mg/kg·day, respectively [7,30-33].

The THQ value is then compared with the acceptable THQ (ATHQ) value to determine whether the risk is acceptable. The ATHQ value depends on the level of risk that is acceptable for the specific metal and the population being evaluated. If the THQ value is greater than the ATHQ value, then the risk is considered unacceptable, and actions must be taken to reduce the exposure to the contaminant. The hazard index (HI) is used to estimate the overall non-carcinogenic risk to human health from exposure to multiple pollutants. It is the total hazard quotient of all heavy metals determined in the spices, as shown in Equation 6.

$$\mathrm{HI} = THQ(Cu) + THQ(Zn) \tag{6}$$

If the ratio of THQ/HI is greater than or equal to 1, there is a possibility of adverse effects on the consumer, while a value of THQ/HI less than 1 indicates that the consumer is relatively safe from experiencing adverse health effects [34,35].

Table 1. Physicochemical characteristics of soil samples (pH, EC and OM) with mean±SD, n = 3.

Physicochemical parameter	Korarima soil	Ginger soil	Turmeric soil	Black pepper soil
	Mean±SD	Mean±SD	Mean±SD	Mean±SD
pH	6.57±0.08	5.91±0.01	6.32 ±0.08	5.86±0.05
Electrical conductivity (mS/m)	0.28±0.01	0.49±0.01	0.12 ±0.01	0.12±0.01
Organic matter (%)	20.59±0.07	15.54±0.04	15.38±0.50	13.93±0.06

Table 2. Percent recovery values (%R±SD, n = 3) for plant samples (BDL: Below detection limit).

Samples	Concentration (mg/kg)	Metals						
		Ca	Cd	Cr	Cu	Ni	Zn	Pb
Korarima	CPS [a]	1.72	BDL	BDL	0.37	BDL	0.67	BDL
	AA [b]	0.30	1	4	0.60	4	0.30	4
	ASP [c]	2.00	-	-	0.94	-	0.95	-
	AR [d]	0.28	0.84	3.91	0.57	3.93	0.28	3.87
	%R [e]	93.33±2.3	84±0.1	97.75±0.7	95±5.02	98.25±2.50	93.33±2.1	96.75±1.98
Ginger	CPS [a]	1.63	BDL	BDL	0.27	BDL	0.76	BDL
	AA [b]	0.30	1	4	0.70	4	0.3	4
	ASP [c]	1.93	-	-	0.95	-	1.07	-
	AR [d]	0.30	0.94	3.87	0.68	3.96	0.31	3.94
	%R [e]	100±1.0	94±0.85	98.75±0.1	97.14±5.2	99±3.3	103.33±1.2	98.5±1.98
Turmeric	CPS [a]	1.71	BDL	BDL	0.25	BDL	0.53	BDL
	AA [b]	0.30	1	4	0.70	4	0.50	4
	ASP [c]	2.04	-	-	0.96	-	1	-
	AR [d]	0.33	0.88	3.96	0.71	4.02	0.47	4.12
	%R [e]	110±0.7	88±0.88	99±0.3	101.42±1.8	100.5±0.32	94±0.8	103±1.6
Black pepper	CPS [a]	1.61	BDL	BDL	0.53	BDL	0.32	BDL
	AA [b]	0.30	1	4	0.50	4	0.70	4
	ASP [c]	1.89	-	-	1.06	-	1.05	-
	AR [d]	0.28	0.97	4.01	0.53	4.03	0.73	3.99
	%R [e]	92.5±0.7	97±0.69	100.25±0.2	106 ±1.9	101± 0.94	104.28±0.4	99.75±1.3

[a] Concentration in plant sample.
[b] Amount added.
[c] Amount in the spiked plant sample.
[d] Amount recovered.
[e] Percent recovery.

3. Results and discussion

3.1. Physicochemical characteristics of the soil samples

Soil pH plays an important role in the transformation of metals from immobile solid forms to more mobile forms. The present study indicated that the soil pH was within the range of 5.86±0.06 to 6.57±0.08, which categorizes the soil as weakly acidic. This pH range is likely favorable for spice crops such as cardamom and black pepper [26]. The solubility of heavy metals is generally higher at lower pH levels within the normal pH range of 5.0-7.0 in agricultural soil [26]. The relatively higher pH values of the soil in this study may have contributed to the low transfer of metals from the soil to the plant, as the soil pH is within the normal range required for agricultural soil, as shown in Table 1.

Metal ions may become unavailable to plants because of complexation with organic matter (OM). Carboxyl groups (-COO-) in both solid and dissolved OM can form stable complexes with metals, leading to the increasing opportunity of forming stable metal-OM complexes as the amount of OM in the soil increases. The OM contents in the soil investigated in this study varied between 13.93 and 20.59%, which is sufficient to form stable metal complexes and decrease the availability of metal ions to plants. Typically, plants do not have the capacities to absorb large metal complexes, resulting in a decrease in metal bioavailability. Metal ions also form strong coordination complexes with OM, causing them to be predominantly bound to the OM fraction in the soil. Consequently, soil OM can be the most significant factor in determining metal availability in the soil. Other workers also reported similar observation [36].

The electrical conductivity (EC) of the solution is a numerical representation of its ability to transmit an electric current. This capability is contingent on the presence of ions and their total concentration, as well as the temperature of measurement [28]. The EC of the farmlands of the soil from the study area was found to vary from 0.12±0.01 to 0.49±0.01 mS/m, as shown in Table 1. These findings indicate that the EC of soils in the study area is relatively low, suggesting that there is a limited amount of ions present in the soil.

3.2. Recovery of the analytical method

The evaluation of the accuracy of the optimized procedures was performed by analyzing the digests of the spiked spice and soil samples for their metal contents. The results of the recovery analysis for the spice samples are presented in Table 2, and the percentage recoveries were found to be within the range of 84.0 to 98.3% for korarima, 94.0 to 103.3% for ginger, 88.0 to 110.0% for turmeric, and 92.5 to 106.0% for black pepper spices, which are all within the acceptable range for all metals [37]. On the other hand, the percentage recovery of soil samples ranged from 88.0 to 106.7%, 87.0 to 113.3%, 86.0 to 113.3%, and 93.3 to 107.5% for the underlying soil samples of korarima, ginger, turmeric and black pepper, respectively, as shown in Table 3, and these findings are also within the acceptable range for all metals analyzed [38].

3.3. Level of metals in spice and soil samples

The spice and soil samples were analyzed for seven target analytes (Ca, Zn, Cu, Cr, Ni, Cd, and Pb) using FAAS in triplicate. The results showed that Cd, Cr, Pb, and Ni were below the method detection limit in all the spice samples. Furthermore, Cd and Pb were not detected in any of the soil samples, and the amounts determined for the remaining metals are presented in Tables 4 and 5. It should be noted, from these findings, that all investigated metals were found to be below the standard set by the World Health Organization (WHO) [21].

Investigations carried out on essential metals present in spice plants revealed that Ca was the most prevalent metal, followed closely by Zn, Table 4. Cu, on the other hand, was found to be the most abundant of the micronutrient heavy metals that were tested. In soil samples, Ca was also found to be the most abundant metal, followed by Zn, Ni, Cr, and Cu, Table 5.

Table 3. Percent recovery values (%R±SD, n = 3) for soil samples (BDL: Below detection limit).

Samples	Concentration (mg/L)	Metals						
		Ca	**Cd**	**Cr**	**Cu**	**Ni**	**Zn**	**Pb**
Korarima	CSS [a]	1.76	BDL	1.09	0.97	1.32	0.73	BDL
	AA [b]	0.30	1.00	1.00	0.30	1.00	0.30	4.00
	ASS [c]	2.07	-	2.00	1.29	2.21	1.04	-
	AR [d]	0.31	0.88	0.91	0.32	0.89	0.31	3.67
	%R [e]	103.33±1.0	88±0.8	91±0.29	106.67±1.18	89±0.66	103.33±2.2	91.75±2.0
Ginger	CSS [a]	1.73	BDL	2.28	0.55	1.05	0.64	BDL
	AA [b]	0.30	1.00	1.00	0.50	1.00	0.30	4
	ASS [c]	2.07	-	3.15	1.03	1.98	0.97	-
	AR [d]	0.34	0.87	0.87	0.48	0.93	0.33	3.73
	%R [e]	113.3±0.4	87±0.14	87±0.8	96 ±1.6	93±0.61	110±0.9	93.25±1.03
Turmeric	CSS [a]	1.65	BDL	2.13	0.50	1.07	0.68	BDL
	AA [b]	0.30	1.00	1.00	0.50	1	0.30	4
	ASS [c]	1.92	-	3.09	0.93	2.08	1.02	-
	AR [d]	0.27	0.91	0.96	0.43	1.01	0.34	4.02
	%R [e]	90±0.6	91±0.8	96±0.6	86±3.7	101±0.7	113.33±0.3	100.5±1.70
Black pepper	CSS [a]	1.19	BDL	2.13	0.28	1.13	0.71	BDL
	AA [b]	0.30	1.00	1.00	0.70	1.00	1.00	4
	ASS [c]	1.47	-	3.14	1.01	2.17	1.03	-
	AR [d]	0.28	0.98	1.01	0.73	1.04	0.32	4.3
	%R [e]	93.33±0.4	98±0.3	101±0.57	104.28±0.4	104±1.14	106 ±1.3	107.5±0.3

[a] Concentration in soil sample.
[b] Amount added.
[c] Amount in the spiked soil sample.
[d] Amount recovered.
[e] Percent recovery.

Table 4. Mean concentration (n = 3, x±SD mg/kg) of the metals in plant samples (BDL: Below detection limit).

Metals	Korarima	Ginger	Turmeric	Black pepper	WHO limits
	Mean±SD	**Mean±SD**	**Mean±SD**	**Mean±SD**	**[21]**
Ca	627.15±36.05	965.46 ±17.5	429.01±1.89	5369.67 ±240.18	-
Cu	7.62±0.27	8.02±0.05	8.53±0.02	10.67±0.24	50 *
Cr	BDL	BDL	BDL	BDL	-
Ni	BDL	BDL	BDL	BDL	50 *
Zn	67.70±2.8	67.02±3.75	52.62±3.27	31.88±0.60	100 *
Cd	BDL	BDL	BDL	BDL	21 *
Pb	BDL	BDL	BDL	BDL	100 *

Table 5. Mean concentrations of the metals (n = 3, x±SD mg/kg) in the soil samples (BDL: Below detection limit).

Metals	Korarima soil	Ginger soil	Turmeric soil	Black pepper soil
	Mean±SD	**Mean±SD**	**Mean±SD**	**Mean±SD**
Ca	3533.3±176.54	2438.67±41.04	4147.17±329.25	1195.67±35.85
Cd	BDL	BDL	BDL	BDL
Cr	21.92±0.79	45.76±0.8	42.74 ±1.18	42.78±0.15
Cu	19.59±0.58	13.03±0.68	16.39±0.29	28.67±1.36
Ni	33.15±1.55	26.27±0.62	21.4±0.13	56.93±3.26
Zn	120.25±9.53	112.9±7.17	131.63±7.98	113.8±2.8
Pb	BDL	BDL	BDL	BDL

3.4. Concentration of metals in spice samples

Metals uptake by plants occurs through a variety of complex biochemical processes which are influenced by factors such as the ability of plants to absorb metals from soil, the availability of mineral elements in soluble and absorbable forms, the concentration of specific metals at the site, and the level of soil contamination with heavy metals. As shown in Table 4, all spice samples contained the targeted metals of Ca, Cu and Zn. The metal concentration in korarima, ginger, turmeric, and black pepper was in the following order: Ca > Zn > Cu. Cr, Ca, Ni, and Pb were not detected in any of the spice samples. The metal concentration determined in the spice samples indicated that Ca had the highest concentration and Cu the lowest concentration in all samples from the spice plant. Specifically, the concentration of Ca in korarima, ginger, turmeric and black pepper was 627.15±36.05, 965.46±17.54, 429.01±1.89, and 5369.67±240.18 mg/kg dry weight, respectively. For Cu, the concentrations in the same order were 7.62±0.27, 8.02±0.05, 8.53±0.02, and 10.67±0.24 mg/kg dry weight. The concentration of Zn in korarima and ginger was comparable, at 67.7± 2.8 and 67.02±3.75 mg/kg, respectively, while turmeric contained 52.62±3.27 mg/kg. The lowest quantity of Zn was obtained in black pepper, at the level of 31.88±0.60 mg/kg.

3.5. Concentration of metals in soil samples

There are varying levels of metal concentrations in the root zone of spice plants, as indicated in Table 5. Among the targeted species, Ca, Cu, Zn, Cr, and Ni were found in the root zone of the spice plants, while Pb and Cd were not detected. The mean concentration level in the cultivated soil sample of ginger and turmeric was found to be on the order of Ca > Zn > Ni > Cr > Cu.

The concentration of Ca in soil samples was found to be in decreasing order as follows: ginger soil > korarima soil > turmeric soil > black pepper soil. The highest concentration of Cr was found in the ginger cultivated soil sample, while the minimum amount of Cr was found in the korarima soil samples. Cu was the metal detected the least in all soil samples compared to other metals. The trend for Zn variation in soil samples was in decreasing order as follows: turmeric soil > black pepper soil > ginger soil. The concentration level of Ni was also found to be in decreasing order as follows: black pepper soil > korarima soil > ginger soil > turmeric soil.

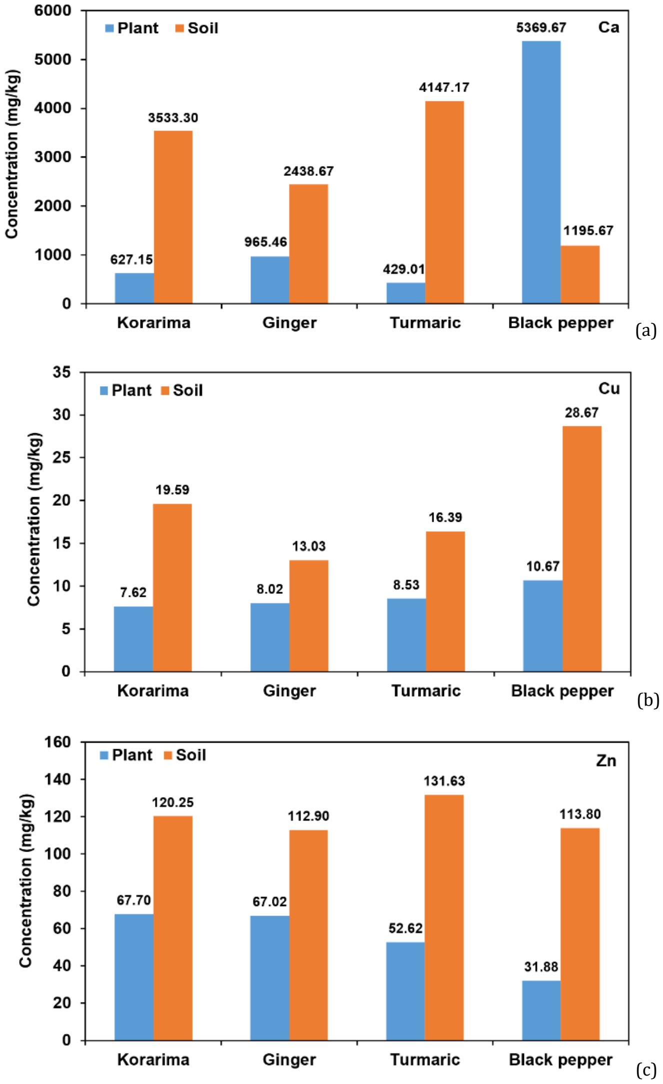

Figure 1. Comparison of metal levels, specifically (a) calcium (Ca), (b) copper (Cu) and (c) zinc (Zn), between korarima, ginger, turmeric, and black pepper plants, along their corresponding soil samples.

3.6. Comparison of metals concentration in plant and soil samples

The detected metal levels were found to be higher in soil samples than in plants grown in those soils, with the exception of black pepper soil, which had lower levels of Ca metal than the black pepper plant. This suggests that only a small portion of the metals in the soil are transferred to the parts of the plant, and the roots act as a barrier to the movement of metals within the plants. The soil samples analyzed were found to contain higher concentrations of all metals than the spice plants, except for black pepper soil for Ca metal, as depicted in Figure 1. This could be due to the fact that the uptake of metals by plants from the soil is significantly influenced by factors such as metal levels in the soil, soil pH, the presence of competing ligands, the ionic strength of the soil solution, the organic matter content of soil type and variety of plants, the age and the simultaneous presence of competing metals [39]. As shown in Figure 1, there is a direct correlation between the levels of metals in the soil and the plant samples, such that an increase in the concentration of metal in the soil leads to an increase in its accumulation in the plant. However, some discrepancies observed for specific metals considered in the study. For example, the concentration of Ca in the soil sample was surprisingly lower than in the black pepper plant. This difference may be attributed to the significant uptake of metal by plants. However, for the other plants, the concentration of metals in the plants was lower than that in their respective soils.

3.7. Translocation factor

The uptake of metals from soil by plant roots and their subsequent distribution to various plant tissues is the phenomenon that is studied using the translocation factor (TF). This index is calculated as the ratio of the concentration of the specific metal in plant tissue to the concentration of the same metal in soil, both represented in the same units. Higher TF values (greater than 1) indicate a higher absorption of metal from the soil by the plant and a higher suitability of the plant for phytoextraction and phytoremediation. On the contrary, lower values indicate a poor response of plants towards metal uptake, and the plant can be considered for consumption. The present study found that the TF values varied between 0.103 and 4.491 for Ca, 0.183 and 0.389 for Cu and 0.280 and 0.594 for Zn, as shown in Figure 2. All reported values in this study were found to be less than one, except Ca in the black pepper plant, which was higher than one.

Table 6. Comparison of the metal concentrations (mg/kg) in spice plants of this study with other studies reported from other parts of the world.

Spice plants	Concentration (mg/kg) of metals in plants								References
	Origin	**Ca**	**Cu**	**Cr**	**Cd**	**Ni**	**Zn**	**Pb**	
Rosemary	Ghana	-	11.0	-	-	35	71	-	[49]
Elettaria cardamomum	Saudi Arabia	-	48.2	-	-	-	50.6	-	[50]
Coriander	India		20.8	0.64	BDL	8.96	134.49	-	[51]
Marjoram	Turkey		11.3	3.2	BDL	3.2	83.7	BDL	]52]
Peppermint	Egypt	-	2.11	-	BDL	0.96	11.4	-	[53]
Rosemary	Turkey	-		-	BDL	-	-	BDL	[52]
Nutmeg	Nigeria		0.01	0.01	0.5	9.3	7.2	5.3	[54]
Ginger	Ghana	-	9.0	-	-	43	73	115	[55]
Red pepper	Canary Island	231	-	-	-	-	-	-	[56]
Korarima	Ethiopia	627.15	7.62	BDL	BDL	BDL	67.7	BDL	This work
Ginger	Ethiopia	965.46	8.02	BDL	BDL	BDL	67.02	BDL	This work
Turmeric	Ethiopia	429.01	8.53	BDL	BDL	BDL	52.62	BDL	This work
Kundo Berbere	Ethiopia	5369.7	10.67	BDL	BDL	BDL	31.80	BDL	This work

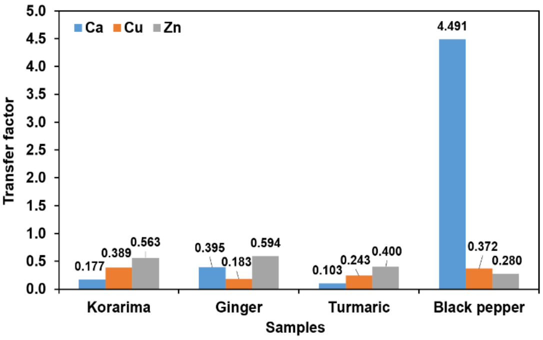

Figure 2. Comparison of the translocation factor (TF) values of the metals in the spices (korarima, ginger, turmeric, and black pepper).

The zinc TF value was the highest in all spice plants (except black pepper). Therefore, it can be concluded that plant species that are nutritionally insignificant to humans absorb higher concentrations of Zn (except for black pepper). TF values for Zn were found to be higher than those for Ca and Cu (except for black pepper), indicating that plants show a higher affinity for Zn than Ca and Cu [40,41].

3.8. Trend of variation of metals in plants

Calcium: Ca concentrations in the fruits of black pepper, rhizomes of ginger, seeds of korarima, and rhizomes of turmeric were 5369.67±240.18, 965.46±17.54, 627.15±36.05, and 429.01±1.89 mg/kg, respectively. These findings indicate that the spice plants studied had higher levels of Ca, which is a highly mobile nutrient that can be translocated to different parts of the plant. Furthermore, plants can easily absorb the wide range of minerals bearing Ca in soil and water samples, as well as its abundance in ground and surface water [42]. Among the plants studied, the black pepper fruits had the highest Ca content, while the turmeric rhizomes had the lowest.

Copper: Cu contents in the assayed spice plant materials ranged from 7.62 to 10.67 mg/kg, with the lowest concentration in korarima and the highest in black pepper. The results obtained are within the permissible limit of 50 mg/kg established by the WHO for edible plants [43]. The concentration of Cu in plant materials is comparable to the reported values for other spice plants [44]. The availability of Cu for plant uptake is influenced by factors such as root intrusion, water and ion fluxes, and the kinetics of metal solubilization in soils, as well as biological parameters such as kinetics of membrane transport, ion interactions, and metabolic fate of absorbed ions [45].

Zinc: The element Zn was found to have a concentration range of 31.88 to 67.7 mg/kg in the investigated plant materials. Among these, the seeds of korarima showed the highest concentration of Zn at 67.7±2.8 mg/kg, which is nearly comparable with the ginger rhizomes at 67.02±3.75 mg/kg. On the other hand, black pepper fruit had the lowest concentration of Zn, *i.e.*, 31.88±0.60 mg/kg. The Zn content of the plant materials studied was compared with the 100 mg/kg limit proposed by the Food and Agriculture Organization (FAO) and WHO for edible plants and it was found that all materials contained Zn below this limit [46]. These results are consistent with the Zn concentrations reported for other spice plants of gingers and black peppers [47].

Cadmium, Chromium, Nickel, and Lead: These metals were not detected in all spice samples investigated in the present study (mainly below the detection limit).

3.9. Comparison of the metal levels of spice plants with values from the literature

The levels of metals in the spices of the farmlands of Southwest Ethiopia were compared with those reported in various regions of the world, Table 6. The results of the current study were generally consistent with most of the values reported, particularly for the essential metals investigated. The Cu concentration in the spices used for comparison ranged from 0.01 to 48.2 mg/kg [48-54], while the value determined in this study fell within that range between 7.62 and 10.67 mg/kg. The mean concentration of Zn was found to be between 7.2 and 134.49 mg/kg in the literature [49-55], while between 31.80 and 67.7 mg/kg in this study. Ca was identified as the most prevalent major metal in all the spices investigated, with an average concentration ranging from 429.01 to 5369.67 mg/kg. Black pepper had the highest Ca content, while turmeric had the lowest. In comparison, the Ca concentration determined in the published literature ranged from 231 to 2540 mg/kg [56].

Most reports in the literature indicate that the levels of Cd and Pb are below the detection limit, which is consistent with the findings of this study and suggests a low exposure to Cd and Pb on farmlands. However, a high value of Pb has been reported in Ghana ginger (115 mg/kg) [55]. The minimal levels of risk for exposure to hazardous metals by oral route are 0.0002 mg/kg per day for Cd and 0.0002 mg/kg per day for Pb as well. Since the human need for spice plants is minimal (in grams per day), there is no risk associated with their use in food flavoring in the current study.

Table 7. EDI, THQ, and HI values of heavy metals in different spices from the Southwest Jimma Zone, Ethiopia.

Spices	Cu		Zn		Hazard index
	EDI	THQ	EDI	THQ	
Korarima	0.00127	0.03175	0.01128	0.03761	0.06936
Ginger	0.00134	0.03342	0.01117	0.03723	0.07065
Turmeric	0.00142	0.03554	0.00877	0.02923	0.06477
Black pepper	0.00178	0.04446	0.005313	0.01771	0.06217

Most of the spices reported in the literature considered for comparison in this study did not have a reported value for Cr. However, values for Cr with a concentration range of 0.01 to 3.81 mg/kg were found in some reports [51,52,54]. In the current study, the concentration of Cr was found to be below the detection limit. The concentration of Ni in the literature ranges from 0.96 to 43 mg/kg, [49,51-55], which is below the WHO allowed limit of 50 mg/kg [21]. In this study, Ni was not detected in the spice plants and may be the detection limit was below the instrument detection limit. Furthermore, the toxicity of Ni is not a common occurrence because its absorption by plants is low. Similar comparisons reported by other workers [57] are also provided as additional literature resource.

3.10. Comparison of metals in soil samples with different standards

The concentration of essential and non-essential metals in the soil samples from the current study was found to vary. Specifically, the observed ranges for Ca were 1195.67-4147.16, Cu: 13.03-28.67, Cr: 21.92-45.76, Ni: 21.40-56.93, and Zn 112.9-131.63 mg/kg. Compared to the international regulatory standards established by Great Britain, which set maximum permissible limits of 100, 50 and 300 mg/kg for Cu, Cr and Zn, respectively, the concentration of Cu, Cr, and Zn in the soil samples studied were found to be lower than the indicated regulatory standard limits [58]. However, the concentration of Ni found in the black pepper soil was slightly higher than the maximum allowed limit of 50 mg/kg, but still lower than the standard set by the WHO of 68 mg/kg [58]. The concentration range of Cu, Cr, Ni, and Zn was below the limit for all investigated soil samples and was in agreement with the standards established by the US Environmental Protection Agency (USEPA) [58]. However, the concentration of Zn in all soil samples was above the limits established by WHO, which is 50 mg/kg [59]. The relatively high Ca concentration range observed in this study may be beneficial for plant growth, since the Ca concentration in the soil is tolerable. Overall, the concentration ranges of metals in the soil samples were within acceptable limits compared to other international standards.

3.11. Pearson correlation of metals in spice plant samples

The relationship between heavy metals in spice plants and their corresponding soil samples is determined by measuring the mean heavy metal content for each sample. A strong positive correlation exists when the correlation coefficient (r) is +1, no correlation when r is 0 and a strong negative correlation when r is -1. The strength of the relationship between the two variables is determined by how close the r value is to +1 or -1. In the korarima plant, there is a good positive correlation between the concentration of Ca and Zn (r = 0.9998), as well as between Cu and Ca (r = -0.99303) and between Zn and Cu (r = -0.9904). Similarly, in ginger, there is a strong positive correlation between Cu and Ca (r = 0.98576). In the turmeric plant there is a strong positive correlation between Cu and Ca (r = 0.86903), as well as between Zn and Ca (r = 0.98702) and between Zn and Cu (r = 0.77829). In black pepper, there is a strong positive correlation between Cu and Ca (r = -0.87369), as well as between Zn and Cu (r = -0.68583). However, there is no significant correlation between Zn and Ca (r = 0.46016) or between Zn and Cu (r = 0.3043) in the ginger, or between Zn and Ca (r = 0.24518) in the black pepper. Furthermore, the correlation coefficients for the metals in the soil samples were also calculated in the same manner.

3.12. Statistical analysis for spice samples

The purpose of this study was to determine whether there were significant differences in the metal content between different spice plants. To achieve this, pairwise statistical analyses were performed using one-way ANOVA, and the results were analyzed using Microsoft Excel®. The findings showed that there were significant differences ($p < 0.05$) in the levels of Ca, Cu, and Zn metals in all spice plants studied at a 95% confidence level. Variations in metal absorption between different plant species can be attributed to various factors such as climate, soil type, and the age of the collected plant materials; in addition, the age of the plant materials can also influence the level of metals present in them.

3.13. Statistical analysis for soil samples

For soil samples where korarima, ginger, turmeric, and black pepper were grown, notable disparities ($p < 0.05$) were observed for Cu, Cr, and Ni at 95% confidence level. However, there was no significant variation ($p < 0.05$) at the 95% confidence interval for Ca and Zn metals between the soil samples where the plants were grown. The presence of metals in the considered soil or disparities may result from improper agricultural practices, including the use of fertilizers, insecticides, irrigation, *etc*. [60, 61].

3.14. Health risk assessment

According to Table 7, the estimated daily intake (EDI) values for copper (Cu) and zinc (Zn) were found to be within the range of 0.00127-0.00178 and 0.005313-0.01128, respectively. These values are comparable to the earlier findings of Tefera and his co-workers, who reported EDI values of 0.0018-0.0270 and 0.0018-0.0090 for Cu and Zn, respectively [62]. Additionally, the target hazard quotient values (THQ) for this study ranged from 0.03175-0.04446 for Cu and 0.03761-0.01771 for Zn. The THQ values in all spices were less than one, indicating that consuming spices does not pose potential health risk to both Cu and Zn [7, 55-57]. Similar results were reported in another study, which found THQ values ranging from 0.0499 to 0.1453 for Cu and 0.0062 to 0.0301 for Zn [62,63]. Furthermore, all the hazard index (HI) values calculated due to the consumption of spices were found to be less than one, suggesting that exposed consumers are unlikely to be at risk of carcinogenic health hazards when the HI values are greater than 1.0 [63].

4. Conclusions

This study was carried out on spices such as korarima, ginger, turmeric and black pepper cultivated in Ethiopia and the soil samples collected from their corresponding sampling sites with the aim of determining the contents of Ca, Cd, Cr, Cu, Ni, Pb and Zn using FAAS. The results showed that Ca, Cu, and Zn metals were detected in the spice plants, while the remaining metals were below the detection limit of the instrument. The highest concentration of Ca was found in black pepper. Soil samples were found to contain Ca, Cr, Cu, Ni, and Zn metals, while Cd and Pb were below the detection limit. The digestion operation used for soil and plant samples was optimized to ensure complete dissolution of the matrices and used a minimum volume of acids, resulting in low blank values, detection, and quantification limits. The optimized digestion procedure was found to be effective, as shown by the excellent recovery for both soil and plant samples. The metal content of the spice plants was within the safe limits set by WHO/FAO, and statistical analysis revealed significant variation in metal levels between spice plants, which could be attributed to differences in the chemical composition of seeds, fruits and rhizomes, environmental conditions or concentration differences in mineral nutrients in the corresponding soils. The findings of the present study suggest that there is no carcinogenic risk associated with the metals present in spices, as indicated by THQ, EDI, and HI. In general, the study reveals a low health risk posed by Cu and Zn metals through the consumption of spices. It is recommended that constant monitoring of heavy metals is carried out in all types of food products to evaluate the potential health risks of toxic heavy metals and protect the health of consumers.

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Ethical approval: All ethical guidelines have been adhered to.
Sample availability: Samples of the compounds are available from the authors.
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Conceptualization: Tura Gemechu; Negussie Megersa; B. S. Chandravanshi; Methodology: Tura Gemechu; Bhagwan Singh Chandravanshi; Negussie Megersa; Software: Tura Gemechu; Abi Legesse; Negussie Megersa; Validation: Negussie Megersa; Tura Gemechu; Bhagwan Singh Chandravanshi; Formal Analysis: Tura Gemechu, Abi Legesse; Negussie Megersa; Investigation: Tura Gemechu; Abi Legesse; Resources: Negussie Megersa, Bhagwan Singh Chandravanshi; Data Curation: Abi Legesse, Tura Gemechu; Writing - Original Draft: Tura Gemechu; Abi Legesse, Negussie Megersa; Writing - Review and Editing: Negussie Megers; Bhagwan Singh Chandravanshi; Visualization: Abi Legesse, Negussie Megersa; Funding acquisition: Negussie Megersa, Bhagwan Singh Chandravanshi; Supervision: Neussie Megersa, Bhagwan Singh Chandravanshsi; Project Administration: Negussie Megersa; Bhagwan Singh Chandravanshi.

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