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Heavy Metals Content of Spices Available on the Market of Asmara, Eritrea

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Authors' contributions

This work was carried out in collaboration among all authors. Authors GK and GA designed the experiment, supervised the study and wrote the manuscript and authors ER, GK and TG carried out the sample collection, lab works and literature review of study. All authors read and approved the final manuscript.

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ABSTRACT

Spices and herbs are being added to diet as ingredients often to improve color, aroma and acceptability of food. The presence of heavy metals in spices could result in the accumulation of these metals in the body organs. The amount of essential and non-essential heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn) in spices commonly used in Eritrea were determined using a dual viewing ICP-OES. Fe was found to have the highest concentration of all the studied metals in all studied spices that ranged from 197-2364 $mgkg^{-1}$. Co was found to be the least accumulated metal in all the species except in Rosemarie and Cinnamon. The level of Fe in all the studied spices (except Cinnamon), the level of Cd in Cumin and Cinnamon, the level of Cu in Allspices, the level of Mn in Black pepper, Allspices, Nutmeg and Cinnamon, the level of Pb in Turmeric and the level of Zn in Nutmeg were found above the WHO Maximum Permissible Limit (MPL). Cumin, Cinnamon, Black pepper and Nutmeg are constituents of the spice called Allspices. Allspices (comprising of almost fourteen spices) is added to berbere which is traditionally prepared and most consumed powder in Eritrea. Cumin was found to be the greatest accumulator of Co, Cr, Ni and Fe. Moreover, the levels of Cr and Cu in Cumin were alarming. Therefore, based on the results of

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this study consumption of Cumin, Cinnamon, Allspices, Black pepper and Nutmeg may have a serious health threat to consumers because berbere is consumed in large amount and with high frequency by Eritrean people. Based on the results of this study, it is recommended that either the consumption of berbere has to be reduced or the addition of spices to berbere has to be controlled.

Keywords: Heavy metals; spices; WHO permissible limits.

1. INTRODUCTION

With the current emphasis on eating more healthy diets that are low in fat and salt, people are turning to various herbs and spices to flavor their food. The culinary herbs and spices that are used to enhance the flavor of vegetables, soups, stir-fry, and pasta dishes can be derived from the bark, buds, flowers, leaves, fruits, seeds, rhizome, or roots of a plant [1]. Spices and herbs belong to condiments, substances which do not contain nutritive components. Although a few dozen different spice plants are of global importance, many more are used as condiments locally, in the regions of their natural occurrence. Some of them are traded in small quantities and used in ethnic restaurants [2]. By clubbing spices and condiments into one group the International Organization for Standardization (ISO) illustrated that the term spice or condiment applies to "such natural plant or vegetable products or mixtures or thereof, in whole or ground form, as are used for imparting flavour, aroma and piquancy to and for seasoning food" [3]. Various vegetal spices are widely applied in human diet all over the world. The most famous include turmeric, cinnamon, red pepper, black pepper, sumac and dried mint [4].

Besides the use of spices as flavors for making color and odor in food, they are also used for their enormous benefits for human health. Most common spices have been documented to possess outstanding microbial, antidiabetic, antiinflammatory, antioxidant and antihypertensive potential [5]. Even though spices are essential they may contain some toxic chemicals as accumulation of heavy metals derived from the surroundings of their production, processing and storage conditions [6]. Moreover, commercial mills may also introduce some amount of metals into the seasonings due to wear and tear of the machinery during use [2]. Although low levels of some heavy metals such as Cr, Fe, Mn, Co, Zn and Cu are considered essential, low levels of other metals such as Cd and Pb can have toxic effects in human biochemical reactions [7]. Exposures to toxic metals are associated with severe health problems with varying degrees of severity and conditions: Kidney problems, neurobehavioral and developmental disorders, high blood pressure and possibly cancer [5]. Since spices and different herbal flavors are frequently and daily used in the diets, determining the level of these elements is significantly important; because they can have a direct impact on human health [4].

Apart from the usual practices of using spices, Eritreans also apply a significant and varied combination of spices in traditionally prepared powdered foodstuffs such as berbere. Although Eritrea imports most of the spices which exist in Eritrean markets today, there is little information available about the safety of these spices with respect to heavy metals.

Since no study has been conducted on assessing the level of heavy metals in food spices of Asmara, this research aimed to detect the levels of heavy metals contamination on the frequently-consumed spices sold in the local markets of Asmara. Also, the levels of investigated metals were compared with recommended levels set by international organizations. Monitoring the levels of heavy metal toxicity in spices would help ascertain the health impact of taking these spices, and provide relevant data on spices in the nation.

2. MATERIALS AND METHODS

2.1 Sample Collection

Samples of common spices were collected directly from local markets in Asmara and identified according to their common name, local name, scientific name, family name and part of the plant used (Table 1). Around 50 grams of each sample was collected from 10 supermarkets in the city from February to March 2018. Samples of each type were then mixed to form a composite representative sample. Samples were kept in polythene bags and stored in a cool dry cardboard before analysis.

2.2 Reagents and Apparatus

Analytical grade chemicals and reagents were purchased from Sigma-Aldrich Company. 65% nitric acid (HNO_3) and 32% Hydro chloric acid (HCl), were used for digestion purposes. Ultrapure-deionized water (18 Ω) was used throughout the study. The glassware were soaked in 3 M HNO_3 for the whole night and washed and rinsed with deionized water to minimize the chances of interferences. All the chemical analyses were conducted under extractor hood and a digital IR Vortex Mixer (S/N296058 made in Italy) was used for mixing of the solutions.

2.3 Preparation and Digestion

In the laboratory, spice samples (except turmeric and allspice) were washed several times first with tap water and then with distilled water, air dried, and finally oven dried at 105°C for 48 hrs. The samples were ground using a stainless steel roller and sieved to obtain 300 µm fractions. Dry ashing method was used to digest the spice samples. In this method, 2.0 g of each powdered spice sample was weighed and transferred to crucibles and placed in a furnace (at 450°C) for 3 hours. After that, the crucibles were taken out of the furnace and cooled. Each sample was transferred to a beaker containing about 60 mL aqua-regia and was placed in hot plate at 100°C until the volume reduced to 40 mL. Each solution was then filtered into a conical flask and diluted to 100 mL with distilled water. Approximately 20 mL from the diluted sample was transferred into a glass test tube arranged in a rack and transferred to the ICP room for trace and heavy metal analysis [8].

2.4 Quality Assurance

Precision and accuracy of heavy metal analysis was assured through repeated analysis of samples against certified reference materials for all the heavy metals. The results were found within ± 2% of the certified value. Quality control measures were taken to assess contamination and reliability of data. Blank and drift standards were run after five determinations to calibrate the instrument. The coefficients of variation of replicate analysis were determined for different determinations for precision of analysis and variations were found to be less than 10%.

2.5 Instrumentation

A dual viewing ICP-OES (Perkin Elmer Optima 8300, made in Singapore) coupled to an ultrasonic nebulizer CETAC 6000AT + (CETAC, Omaha, NE, USA) was employed for the analysis of the trace and other elements. The Windows 7 compatible S/W provided by Perkin Elmer was used to process the spectral data for calculating sample concentrations by comparing light intensities measured at various wavelengths for standard solutions with intensities from the sample solutions. The operating conditions set for the ICP-OES are shown in Table 2.

2.6 Statistical Methods

All the data were statistically analyzed by analysis of variance (ANOVA) as applicable to a completely randomized block design (CRBD) and factorial experimental design (three factors) using STATA version 14. Means were compared using Tukey's pairwise comparison tests. Statistical significance was defined as $p < 0.05$.

Table 1. Scientific and common names of studied spices

Common name	Local name	Scientific name	Family	Used part
Turmeric	Hrud	*Curcuma longa*	Zingiberaceae	Rhizomes
Allspices	Shewate qememat	*Pimenta dioica*	Myrtaceae	Seeds
Cumin	Camun	*Cuminum cyminum*	Umbellifeae or Apiaceae	Seeds
Black Pepper	Tselim Berbere	*Capsicum nigrum*	Piperaceae	Seeds
Spinach rhubarb[a]	Meqmeqo[a]	*Rumex abyssinicus*	Polygonaceae	Rhizomes
Spinach rhubarb[b]	Meqmeqo[b]	*Rumex abyssinicus*	Polygonaceae	Rhizomes
Nutmeg	Korerima	*Myristica fragrance*	Myristicaceae	Bulb
Cinnamon	Qarfa	*Cinnamonum zylanicum*	Lauraceae	Bark
Rosemarie	Azmarino	*Rosmarinus officinalis*	Lamiaceae	Leaf

Spinach rhubarb[a] = Locally grown; Spinach rhubarb[b] = Imported

Table 2. The operating conditions of the ICP-OES

Condition	Setting
Power	1.3 kW
Plasma gas glow	15 L/min
Auxiliary gas flow	1.5 L/min
Spray chamber type	Glass cyclonic (single-pass)
Torch	Standard one-piece quartz axial
Nebulizer type	Sea spray
Nebulizer flow	0.7 L/min
Pump speed	2-4 rpm
Total sample usage	2 mL
Replicate read time	5 s
Number of replicates	3
Sample undertake delay time	15 s
Stabilization time	40 s
Rinse time	20 s
Fast pump	Off
Background correction	Fitted

3. RESULTS AND DISCUSSION

The results of trace and heavy metal analysis in Turmeric, Allspices, Cumin, Black pepper, Nutmeg, Cinnamon, Rosemarie, and Spinach rhubarb available in and around Asmara markets are given in Table 3. For comparison, the Maximum Permissible Limit (MPL), as applied to other food and condiments based on WHO/FAO standards are also given in the table. Maximum limits for trace and toxic heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) in spices was obtained from [9,10] and that of Co was obtained from [5].

3.1 Cadmium (Cd)

The level of Cd in the studied spices (Table 3) ranged from 0.03 ±.01 in Black pepper to 0.59 ±.01 mg Kg^{-1} in Cinnamon. As depicted in Table 3, Cd content in nearly 78% of the studied spices was found to be within the MPL (0.2 mg Kg^{-1}) except the Cd level in Cumin (0.22 mg Kg^{-1}) and Cinnamon (0.59 mg Kg^{-1}). This high level of Cd might be due to the use of cadmium containing fertilizers or from the practice of growing these crops on the soil amended with sewage sludge [6]. The high levels of Cd may also be associated with the plant and the soil properties (such as calcareous nature of soil) that affect uptake of the metal [7]. High value of Cd in the body induces kidney injuries, leading to aminoaciduria, glycosuria and renal tubule necrosis [11].

As was discussed, Cd levels in Cinnamon and Cumin were found above the MPL. In Eritrea, Cumin, being a constituent of the spice called Allspices, is used in a traditionally prepared popular powder called berbere. Cinnamon is also a constituent of Allspices and is used in small amount in tea and in rice. The major constituent of berbere is pepper and the spice called Allspice (comprising of almost fourteen spices) is added to berbere. Berbere is the most consumed powder in Eritrea added to the majority of soups prepared in Eritrea. It is added to greater extent in soups such as sauce (traditionally called silsi), chicken stew and beef stew (traditionally called zigni). Therefore, based on the results of this study, high level of Cd in Cumin and Cinnamon may have a series health threat to consumers because berbere is consumed in large amount and with high frequency by Eritrean people.

3.2 Cobalt (Co)

Although cobalt is toxic at elevated levels, it is an essential trace element being a component of vitamin B12 precursor and applicable in treatment of anemic patients [1,7]. In the studied spices, cobalt concentrations (mg kg^{-1}) ranged from 0.32±.01 in Cinnamon to 1.95±.13 in Cumin (Table 3). Cobalt levels in all the studied spices were found to be with in the MPL (3.5 mg Kg^{-1}). Therefore, the levels of cobalt determined in all the spice groups may not pose any potential risk. Allspices, Nutmeg and Cumin have relatively high values of Co and thus they can be used as good source of Co for consumers.

Table 3. Mean ± Standard deviation of trace and heavy metal concentrations in studied spice samples in mg/kg

Spices	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Turmeric	0.11±.01	0.69±.03	0.85±.13	10.25±.32	446.26±3.37	41.78±.01	1.12±.02	12.0±1.05	32.26±1.69
Allspices	0.15±.01	0.98±.04	2.00±.35	20.47±.09	1255.2±9.72	137.17±1.79	6.05±.14	4.98±.53	38.67±1.32
Cumin	0.22±.02	1.95±.05	26.63±.18	16.27±0.58	2364.1±7.49	81.81±.29	14.1±.03	5.09±.64	33.77±1.28
Black pepper	0.03±.01	0.58±.02	1.82±.09	10.21±.15	359.32±9.34	197.31±.20	3.76±.02	5.79±1.90	7.35±.10
Spinach rhubarb[a]	0.07±.01	0.56±.03	1.01±.07	4.05±.07	388.05±5.03	19.37±.24	1.15±.12	ND	19.08±.13
Spinach rhubarb[b]	0.11±.01	0.71±.07	1.37±.03	2.56±.16	493.20±8.29	23.52±.93	1.02±.04	2.77±.53	24.58±.61
Nutmeg	0.18±.02	1.10±.02	1.93±.20	11.04±1.29	878.54±6.02	263.24±3.96	2.29±.09	1.58±.12	91.83±.89
Cinnamon	0.59±.01	0.32±.01	0.71±.04	5.34±.01	197.04±3.05	296.16±.62	0.97±.14	ND	10.23±.02
Rosemarie	0.09±.00	0.46±.01	1.96±.08	5.73±0.49	629.00±7.70	20.97±.46	2.08±.12	ND	35.55±1.30
MPL	0.2	3.5	30	20	300	100	50	10	50

ND = Not Detected, Superscript a = grown locally, Superscript b = imported
MPL= Maximum Permissible Limit

3.3 Chromium (Cr)

Exposure to Cr can occur through food, it is considered an essential metal for carbohydrate and lipid metabolism [5]. The level of Cr in the studied spices ranged from 0.71±.04 in Cinnamon to 26.63±.18 mg Kg^{-1} in Cumin. Cr content in all the studied spices (Table 3) was found to be within the MPL (30 mg Kg^{-1}). As discussed, Cd level in Cumin was found above the MPL. Moreover, the Cr level in Cumin is alarming and this shows possible Cd and Cr hazard with the consumption of the spice Cumin because it is a condiment added to berbere. While Cr (III) is essential in human body for the improvement of glucose tolerance, excessive intake, especially of the more oxidizing Cr (VI), can harm biological systems [7]. Chronic exposure to Cr may result in liver, kidney and lung damage, it is toxic only at high levels [1,12].

3.4 Copper (Cu)

Copper plays an important role in growth of bone, connective tissue, brain, heart and many other organs [4]. On one hand Cu plays a role in the oxidative defense system and on the other hand chronic Cu toxicity can result in severe poisoning [6]. The level of Cu in the studied spices ranged from 2.56±.16 in Spinach rhubarb[b] to 20.47±.09 mg Kg^{-1} in Allspices. Cu contents in all the studied spices (Table 3), except Allspices, were found to be within the MPL (20 mg Kg^{-1}) but the level of Cu in Cumin (16.27 mg Kg^{-1}) was also substantially high. The high level of Cu in Allspices and Cumin demonstrates possible Cu hazard because they are constituents of berbere. On the other hand, Turmeric, Black pepper and Nutmeg can be used as good sources of Cu for consumers because they have relatively high values of Cu.

3.5 Iron (Fe)

Iron is the most important element for producing blood cells in the body in a way that 70% of iron is inside blood cells and in the form of hemoglobin with a vital role in transferring oxygen to the human tissues [4]. The level of Fe recorded in the studied spices is relatively very high when compared to the level of the other metals in the spice samples. Iron content of the studied spices (Table 3) ranged between 197.04±3.05 for Cinnamon and 2364.1±7.49 mg kg^{-1} for Cumin. With the exception of Cinnamon, Fe contents in nearly 89% (8 of 9) of the studied spices exceeded the MPL (300 mg Kg^{-1}). The high levels of iron could be due to contamination during milling. Research indicates that grinding of spices in commercial mills contaminates them to about between 3 and 5 folds, due to wear and tear of the machine parts [2]. Iron intake from spices thus has a series effect on the health of consumers. In addition to Cd and Cr, the level Fe in Cumin was also found highest among the studied spices. On the other hand except Cumin and Allspices, in which their Fe level is magnified, the remaining spices can be used as good sources of Fe especially for anemic individuals.

3.6 Manganese (Mn)

Manganese in the body helps building connective tissues, bones, bleeding clotting factors and sex hormones. Manganese has a vital role in calcium absorption, regulating blood sugar and the metabolism of carbohydrates and fats. It is also vital for the natural performance of brain and nerves [4]. Although considerably lower than that of Fe, the level of Mn in the studied spices is also relatively high when compared to the level of the other metals in the spice samples. Mn content (mg kg^{-1}) of the studied spices ranged between 19.37±.24 for Spinach rhubarb[a] and 296.16±.62 for Cinnamon (Table 3). The level of Mn (mg kg^{-1}) in nearly 44% of the spices, (Allspices (13.17±1.79), Black pepper (197.31±.2), Nutmeg (263.24±3.96) and Cinnamon (296.16±.62)), exceeded the MPL (100 mg Kg^{-1}). A deficiency of manganese causes diseases and an excess causes poisoning of the central nervous system [12]. Allspices is the most important constituent of berbere in Eritrea. Black pepper, Cinnamon and Nutmeg are in turn constituents of Allspices. Therefore, high level of Mn in Allspices, Cinnamon, Black pepper and Nutmeg can pose potential health hazard to consumers [13]. Cumin has relatively high values of Mn, and hence can be used as a good sources of Mn for consumers.

3.7 Nickel (Ni)

Nickel is a necessary element for iron absorption and preventing anemia in the body. This element is essential in making enzymes such as dehydrogenases, transaminases, and alpha amylases that handle many chemical reactions of the body. Also, nickel plays a main role in absorbing calcium in the bones [4]. Ni content (mg kg^{-1}) of the studied spices ranged between 0.97±.14 for Cinnamon and 14.1±.03 for Allspices (Table 3). Cumin was found to contain highest level of Ni among the other spices. The content of Ni in all the studied spices was found to be much less than the MPL (50 mg Kg^{-1}). Nickel toxicity is not very common occurrence as its absorption by the body is very low [6,12].

3.8 Lead (Pb)

Lead content (mg kg^{-1}) of the studied spices ranged between zero readings in Spinach rhubarb[a], Cinnamon and Rosemarie to 12.0±1.05 in Turmeric. The content of Pb in nearly all the studied spices except Turmeric (Table 3) was found to be within the MPL (10 mg Kg^{-1}). The relatively high levels of Pb in Turmeric might have resulted from accumulation of Pb through air pollution, inclusion or absorption at the mill during grinding, and from some agrochemicals comprising pesticides, such as Lead Arsenates, applied during cultivation [4,14]. Lead is the most recognized toxic environmental pollutant. It reacts or complexes with many biomolecules and adversely affects the reproductive, nervous, gastrointestinal, immune, renal, cardiovascular, skeletal and muscular systems as well as developmental processes [14]. Even though the level of Pb in Turmeric is higher than the MPL, it may not be a potential health threat to consumers since Turmeric is used in rice and in alcha (a traditionally prepared food containing potato and cabbage) which are prepared in special occasions only.

3.9 Zinc (Zn)

Zinc is an important essential metal in protein synthesis, energy production, and in maintaining the membranes [12]. Zn content (mg Kg^{-1}) of the investigated spices ranged from 7.35±.10 in Black pepper to 91.83±.89 in Nutmeg. With the exception of Nutmeg (91.83±.89 mg Kg^{-1}), the zinc values in the investigated spices are within the tolerable limits (50 mg Kg^{-1}). High level of Zn in Nutmeg may also pose a series health threat to consumers because Nutmeg is a constituent of Allspices and hence berbere. Zinc has a determining role in the performance of over 300 enzymes in the body, participating in their building or regulating their activities. However, high values of zinc in the body interfere with the metabolism and activities of other metals [4]. Conversely, the level of Zn in Turmeric, Allspices, Cumin, Spinach rhubarb[b] and Rosemarie is relatively high and thus they can be used as good sources of Zn for consumers.

3.10 Results of Statistical Analysis (ANOVA)

Results from a two-way ANOVA (RCBD) revealed that the average concentrations of the metals are statistically different ($p < 0.05$) for the different metals. Tukey's Pairwise comparisons also revealed that the average metal concentrations in Cumin, Allspices and Nutmeg are not significantly different among themselves but are significantly different from the average metal concentrations of the remaining spices. Moreover, Tukey's Pairwise comparisons indicated that the average concentration of Fe is different from the average concentrations of the other metals in all the spices.

4. CONCLUSION

The study revealed that Allspices, Cinnamon, Black pepper and Nutmeg were found to concentrate Mn above the WHO permissible limit. Cumin and Cinnamon were also found to concentrate the non-essential element Cd above the WHO permissible limit as do Turmeric for Pb, Allspices for Cu and Nutmeg for Zn. The levels of Cr and Cu in Cumin were also alarming. However, Spinach rhubarb[a], Spinach rhubarb[b] and Rosemarie were found to contain all the investigated metals, except Fe, below the WHO permissible limit. Tukey's Pairwise comparisons revealed that the average metal concentrations in Cumin, Allspices and Nutmeg are significantly different from the average metal concentrations of the remaining spices. Therefore, depending on their origin and the frequency of usage the consumption of the investigated spices is likely to pose different degrees of threats to consumers. Berbere, which comprises the spices with high concentration of metals, is consumed in significant amount and with high frequency in Eritrea; thus it is recommended that the addition of spices to berbere has to be reduced. The samples investigated in this study are composite. However, limitations such as the dilution effect have not been taken into account. Thus, it is suggested that further study be conducted for individual sample of each spices based on the origin of cultivation in order to determine which hotspot is responsible for high level of contamination.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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