

# Metals Contents in Spices and Herbs Available on the Egyptian Market: Assessment of Potential Human Health Risk

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**Abstract:** Spices and herbs are being used as a diet ingredient often to improve color, aroma and acceptability of food. The contamination of spices with heavy metals may result in the accumulation of these metals in the body organs. The aim of the present study was to investigate As, Cu, Fe and Zn contents in the most popular spices and herbs that are used in Egyptian cuisine. Ten samples were purchased directly from the local markets in Egypt. Arsenic, Copper, Iron and Zinc contents in the samples were determined by atomic absorption spectrophotometer after acid digestion. The concentration ranges for the studied elements were found as 0.02-0.11, 3.72-21.04, 47.39-1261.21, and 2.14-314.05mg/kg for arsenic, copper, iron, and zinc, respectively. The obtained data compared well with the counterpart data reported internationally. The World Health Organization has given the maximum permissible limits of arsenic, copper, Iron and Zinc in species, i.e. 1, 50, 300 and 100 mg/kg, respectively.

**Keywords:** Atomic Absorption Spectroscopy, Heavy Metals, Herbs, Spices.

## INTRODUCTION

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 [1]. "Moreover, in the last three decades, mainly because of their medicinal values, the use of spices and other herbs have increased markedly in most regions of the world, including Europe and North America. For instance, during this period, herbal medication in the USA has grown into an industry worth an average of \$1.5 billion per year, with projected annual growth of 15%" [2]. On the other hand, the process of preparation and handling can make them as a source of food poisoning [3]. These spices may easily be contaminated by heavy metals from the soil or aerial depositions as these spices are dried on the ground or on roof tops. Moreover, commercial mills used may also introduce some amount of metals into the seasonings due to wear and tear of the machinery [4].

"Heavy metals are potential environmental contaminants with the capability of finding their way into the food we eat and causing human health problems. They are given special attention throughout the world due to their ubiquitous nature and toxic effects even at very low concentrations" [5]. The accumulation of heavy metals can "have middle term and long term health risks, and strict periodic surveillance of these contaminants is therefore advisable" [6]. As arsenic is

an element, it is not degraded in the environment. Instead, it may accumulate throughout the food chain. Soil, air, food and water are sources of arsenic [7]. The high level of Arsenic can be due to the use of certain fertilizers and pesticides. Higher concentration of Arsenic in food/spices can cause harmful effect on the skin, lungs, liver and bladder. Its lower effects can cause nausea, vomiting or even damage to the blood vessels [8]. "Copper, which is an element essential for many biological systems, plays an important role in carbohydrate and lipid metabolism. Lack of the recommended dietary allowance for copper may be hazardous to human health" [9]. "The analysis of copper levels in food is of special interest to researchers for these reasons" [10]. Fe is an essential element. It is a constituent of active site of various reproductive hydrogenases, most frequently associated with sulfur containing ligands. Fe together with hemoglobin and ferredoxin plays a central role of metabolism. Fe facilitates the oxidation of carbohydrates, proteins and fat to control body weight which is an important factor in some diseases (diabetes) [11]. "Zinc is an element found virtually in every cell of the human body and plays vital roles in the development and healthy growth of the human body. Zinc plays an important role in growth and has a recognized action on more than 300 enzymes by participating in their structure or in their catalytic and regulatory action" [12].

However, there is limited data on the levels of these metals in these plants from the Egyptian markets. This work therefore seeks to bridge that gap by providing information on the levels of heavy metals of these most consumed plants. Also, the levels of investigated metals were compared with recommended levels by the international organizations.

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## MATERIALS AND METHODS

### Sampling and Sample Preparation

Eight different powdered samples were purchased at random from local markets in Egypt. The glassware containers used for analysis in this work were washed with detergent and rinsed several times with tap water to remove any contamination due to detergent; then they were soaked overnight in 6 N HNO<sub>3</sub> (Merck) solutions and finally rinsed several times with deionized water.

The samples were carefully opened and dried to constant weight. 1g sample was digested with 20mL of 2:1 HNO<sub>3</sub>/HClO<sub>4</sub> (Anal grade), and heated until the evolution of white fumes. Where necessary more acid mixture was added and the sample digested until the evolution of white fumes marking the end of the digestion process. The digests were filtered into standard 50mL volumetric flask and made up to the mark with distilled water.

### Analytical Quality Control

"Three replicate digestions were made for each sample. The average of blank signals was subtracted from analytical signals of digested samples". The accuracy and precision of the digestion procedure was carried out in cumin samples by spiking the analyzed samples with aliquots of metal standard and then reanalyzing the samples. All the spiked samples were digested following the procedure described earlier in triplicates. Percentage of recoveries obtained was 90.05, 91.70, 95.4 and 93.76 % for As, Zn, Fe and Cu, respectively and the relative standard deviations (RSDs) of replicates were less than 10% (4.9, 3.5, 4.45 and 3.78) for As, Zn, Fe and Cu, respectively. The detection limit of the method for each metal was As 0.006 mg/kg, Cu 0.029 mg/kg, Fe 0.048 mg/kg and Zn 0.035 mg/kg.

### Instrumentation

AAs measurements were performed with a SHIMADZU AA6650 atomic absorption spectrophotometer equipped with a deuterium background corrector. An atomizer with an air/acetylene burner head was used for determining all the investigated metals. "All instrumental settings were those recommended in the manufacturer's manual book". The wavelengths (nm) used for the determination of the analytes were as follows: As (193.7), Cu (324.8), Zn (213.9) and Fe (248.3). A standard solution for each metal under

investigation was provided by suitable internal chemical standards (Merck Chemicals, Germany). Instruments were calibrated using sets of at least five standards covering the range of concentrations encountered in the literature. All calibration curves showed good linearity ( $r > 0.99$ ). Arsenic was measured based on the following Hydride generation conditions: quartz cell, 16 cm path length, 90.7 cm i.d., electrothermal heating, cell temperature, 900°C, carrier gas (argon) flow rate. 50-100 mL/min. reducing agent (0.2% (w/v) NaBH<sub>4</sub> in 0.05% (w/v) sodium hydroxide solution) flow rate, 5-7 mL/min, and HCl flow rate, 9-11 mL/min [13] using Shimadzu Hydride Vapor Generator model: HVG-1.

## RESULTS AND DISCUSSION

As, Cu, Fe and Zn analyses have been performed on eight common spices and herbs available at local markets in Egypt and the outcome has been presented in Table 1. The values of metal concentration were compared with maximum permissible standard concentration of 1, 50, 300 and 100 mg/kg respectively as recommended by the World Health Organization (WHO) [14] and with the counterpart data reported internationally (Table 2).

### Arsenic

The concentration of arsenic in the investigated samples ranged from 0.02 in paprika, to 0.11 mg/kg in cloves (Fig. 1). The concentrations of arsenic in all the analyzed samples are well within the permissible limits set by WHO [14] (1 mg/kg). It was very well below the limit and may be considered tolerable.

### Copper

Results in Fig. (2) showed that the highest value of 21.04 mg/kg for Cu was registered for hot red pepper, while paprika recorded the lowest value of 3.72 mg/kg. All the investigated samples recorded values which were lower than half the WHO limit for Cu in the spices. "Therefore, the Cu levels of the samples are relatively very tolerable. Ozkutlu *et al* [15] reported a range of 3 to 11 mg/kg for Cu levels in some spices while a previous work reported 6-17 mg/kg" [16].

### Iron

"Thus Iron has a relatively high WHO level in foods". However, hot red pepper, cloves, parsley and black pepper had Fe levels of, 1361.21, 1180.54, 1230.18 and 452.13

**Table 1. Levels (mg/kg) of selected metals in spices samples consumed in Egypt.**

As		Cu	Fe	Zn
Min	0.02	3.72	47.39	2.14
Max	0.11	21.04	361.21	314.05
Mean	0.05	8.27	620.02	132.98
SD	0.03	5.73	541.36	133.91
Permissible limits WHO (1989)	1	50	300	100

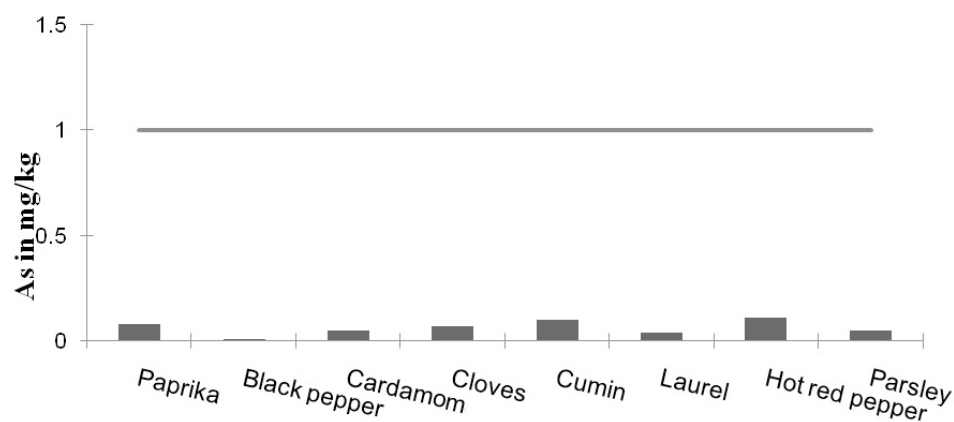


Fig. (1). Levels of Arsenic in different spices.

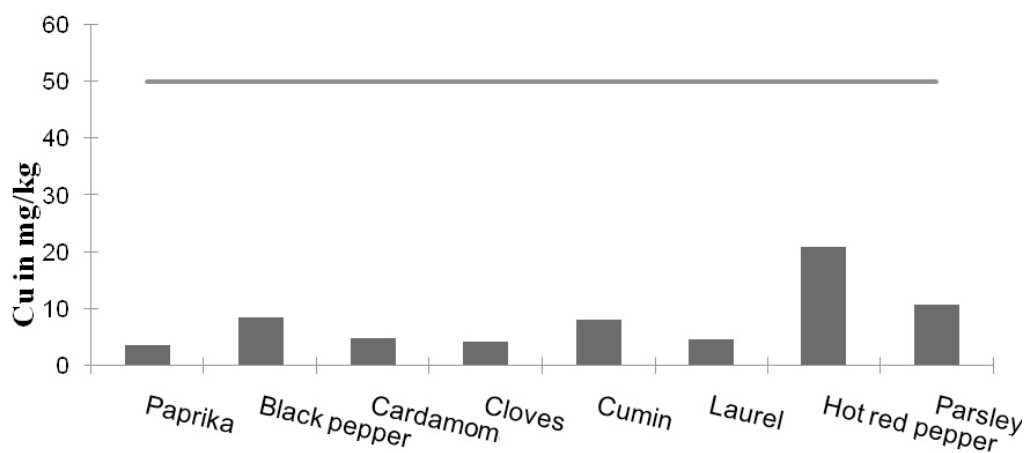


Fig. (2). Levels of Copper in different spices.

Table 2. International comparison of the data.

Reference	As (mg/kg)	Cu (mg/kg)	Fe (mg/kg)	Zn (mg/kg)
This work	0.02-0.11	3.72-21.04	47.39-1361.21	2.14-314.05
[1]	NA	4.2-9.12	NA	6.86-30.84
[4]	NA	0.9-10.10	19.4-971.4	2.4-34.60
[8]	0.7-1.5	2.3-19.69	NA	NA
[21]	NA	3.8-35.4	30-945.3	5.2-83.7
[22]	NA	9-44	144.5-1260	NA
[23]	NA	10.5-22.5	56-650	6.1-47
[24]	<0.07-0.37	NA	NA	NA
[25]	NA	8.88-32.62	115.25-1265.76	33.90-96.55
[26]	ND-0.9	NA	NA	NA

NA; Not available  
ND, Not detected

mg/kg respectively, which are distinctly beyond the WHO limit. 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 [17]. On the other hand, cumin, laurel, cardamom and paprika Fig. (3). may be considered as alternate sources of Fe, "as

they recorded levels below that of the WHO but were yet rich in Fe, as far as this study report is concern".

#### Zinc

In case of zinc, there was a large variation in concentration for all the samples ranging from 2.14 mg/kg for paprika and 314.05 mg/kg for cumin (Fig. 4). The

obtained results showed higher values than those values (59-74) and (7.2-16.3) mg/kg reported by Nkansha and Amoako [18] and Iwegbue *et al.* [19] respectively. "It is therefore imperative to perform routine monitoring on the levels of Zn in these well-patronized spices so as to avert encountering prolific zinc toxicity"[18].

The relatively high level of the essential elements iron (Fe) and zinc (Zn) in this work corroborates the result of Onianwa *et al.* [20] "and reflects the normal composition expected of plant-derived products, which most of the samples are. Plant foods and plant derived foods contain iron in the form of metalloproteins, plant ferritins, iron present in the sap and iron complexed to structural components or storage compounds predominantly as phytates".

**CALCULATION OF THE DAILY DIETARY INTAKE**

Heavy metals and As are the subjects of many studies because of their toxicity, persistence in the environment and bioaccumulative ability. Indeed, they are accumulated through food chains up to toxic concentrations; therefore, they can pose an adverse potential threat both for animal and human health [27].

As consumption of spices is a possible source of heavy metals accumulation in human body, there is a great interest in estimation of the daily dietary intakes of these metals through spices consumption.

The daily dietary intake (mg/day/person) was calculated based on these suppose:

- 1) The human weight is 50 kg
- 2) The human intake from spices per day is 20 g

Where

$$\text{The daily dietary intake (mg/day/person)} = A \times B$$

**A** is the average quantity of spices in gram consumed per individual per day

**B** is the concentration of metal (mg/kg) in studied samples [22].

The recommended dietary allowances (RDA) as mg/day/person for copper, iron and zinc are 2, 18 and 15, respectively [28]. The daily dietary intake values calculated for spices examined in present study were lower than the Food and Nutrition Board of the National Academy of Sciences, United States recommendations given above.

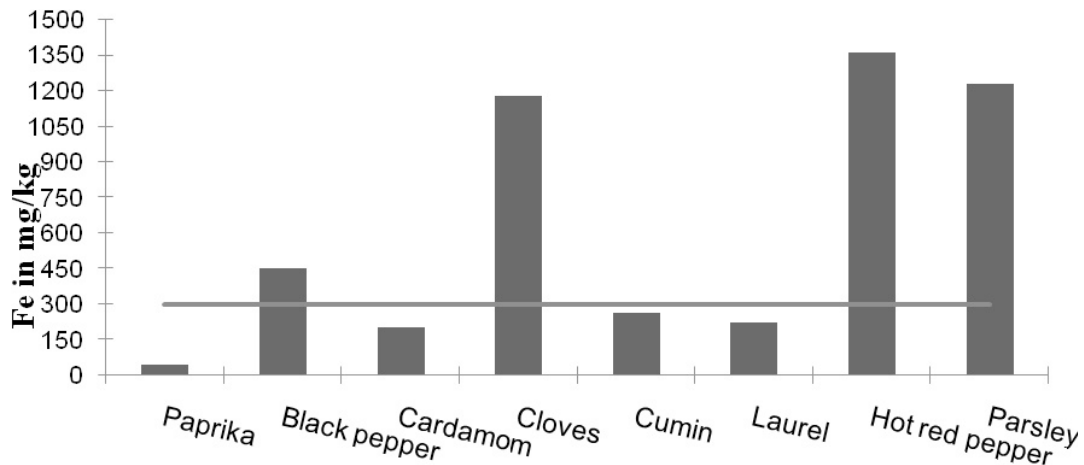


Fig. (3). Levels of Iron in different spices.

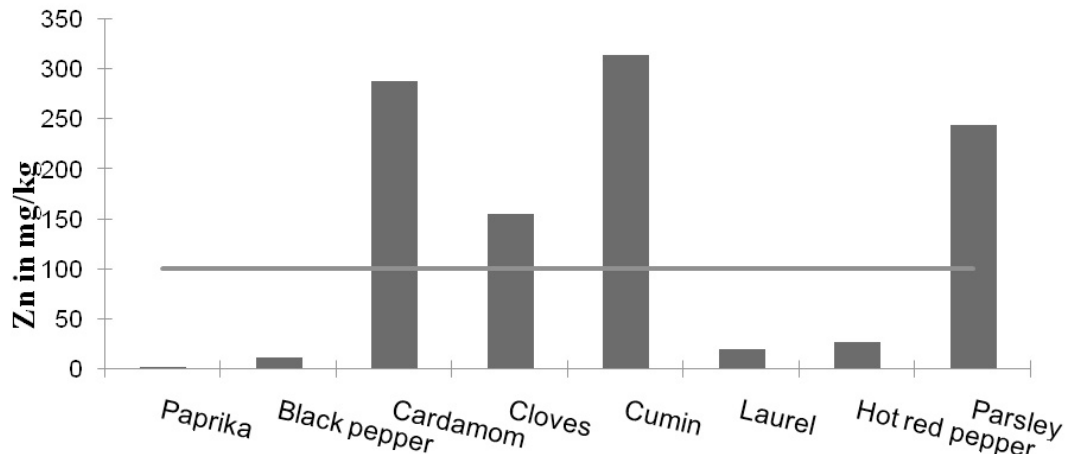


Fig. (4). Levels of Zinc in different spices.

**Table 3. Target hazard quotients (THQs) and total target hazard quotients (TTHQs) for metals by consumption of spices.**

TTHQs	THQs				Sample
	As	Cu	Zn	Fe	
0.08	0.014	0.037	0.003	0.027	Paprika
0.39	0.034	0.086	0.008	0.258	Black pepper
0.39	0.027	0.049	0.196	0.115	Cardamom
0.90	0.075	0.043	0.106	0.675	Cloves
0.47	0.020	0.082	0.214	0.152	Cumin
0.22	0.027	0.048	0.014	0.126	Laurel
1.03	0.020	0.210	0.018	0.778	Hot red pepper
1.01	0.034	0.107	0.166	0.703	Parsley

The weekly tolerable intake value of As is 7µg/kg body weight [29]. On the other hand it is stated 12-25 µg/day As is enough to meet the requirements [30]. It is not possible to provide As by consuming only these species.

#### HUMAN RISK ASSESSMENT ANALYSIS

The health risks from consumption of spices were assessed based on the target hazard quotient (THQ). The THQ is a ratio of determined dose of a pollutant to a reference dose level. If the ratio is less than 1, the exposed pollution is unlikely to experience obvious adverse effects. The method of estimating risk using THQ was provided in the USEPA region III risk-based concentration table (USEPA 2007) [31] and it is described by the following equation:

$$THQ = \frac{EF \times ED \times FI \times MC}{RfD \times BW \times AT}$$

Where THQ target hazard quotient; EFr is the exposure frequency (365 days/year); ED is the exposure duration over a lifetime (70 years), FI is the mean ingestion rate (20g/person/day); MC is the metal concentration in spices (µg/g, on wet weight), and AT is the average life time (70 years x 365 days/year); RfD of the oral reference dose for Fe, Cu, Zn, and As suggested by the USEPA (2011) [32] 0.7, 0.04, 0.3 and 0.0003µg/g/day, respectively; BW is the average body weight (50 kg); AT is averaging time for non-carcinogenes (365 days/year x number of exposure years, assuming 70 years in this study).

The THQ has been recognized as a useful parameter for the evaluation of risk associated with the consumption of metal contaminated food [33]. The target hazard quotients (THQs) values of As, Cu, Fe, and Zn for the investigated samples do not exceed one indicating that there is no health risk from consuming these spices (Table 3). However, the total target hazard quotients (TTHQs) ranged from 0.08 to 1.03 for the consumption of spices (Table 3). The TTHQs value caused by consuming hot red pepper (1.03) and parsley (1.01) were quite higher than 1, and likely will pose a potential risk to the local inhabitants through its consumption.

#### CONCLUSION

Metals are present in spices and herbs at different concentrations, which in some cases exceeded the permissible levels. Grinding of spices in commercial mills contaminates them due to wear and tear of the machine parts. In general, routine check and frequent analysis of foodstuff is required to avoid the risk of exceeding the intake beyond the tolerance limits standards. Moreover, the results of this work can also be used to understand the chemical quality of spices, and to evaluate the possible risk associated with their consumption.

#### CONFLICT OF INTEREST

The author confirms that this article content has no conflict of interest.

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