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Evaluation of Some Metals in Commonly Consumed Spices in Bahrain

Abstract: Concentration of some heavy and essential metals were assayed in seventeen commonly consumed spices in Bahrain using spectroscopy atomic absorption. Samples were collected from different retail outlets in the local spice market (bazaar). The data showed wide variation in metal contents among the various spice samples. The maximum mean level of elements among all spices based on plant parts fall in the magnitude of the order: iron> zinc > copper > nickel > lead > cadmium, in leaves, rhizomes, seeds, buds, fruits and barks, respectively. For heavy metals, caraway contained the highest level of lead (2.2 µg/g) and green cardamom exhibited the highest cadmium level (0.9 µg/g). With essential metals, concentration of iron, zinc, copper and nickel were highest in cumin (13.6 µg/g), black cumin (52.2 µg/g), black pepper (17.3 µg/g) and black cumin (4.9 µg/g), in that order. The reliability of the findings and approach was confirmed by analyzing data from the literature.

Keywords: accumulation, Bahrain, condiments, toxic metals, heavy metals, spices

تقييم لمحتوى المعادن في التوابل الشائعة الاستعمال في البحرين

عواطف مهدي حسن ، قاهر على منديل و حسين عبدالنبي

المستخلص : تتناول هذه الدراسة، تركيز بعض المعادن الأساسية والثقيلة في سبعة عشر نوع من التوابل الشائعة الأستعمال في البحرين .جمعت العينات من موزعين محليين للتوابل، وتم تجفيفها وطحنها وتحليل المستخلص السائل المذاب، في وسط حمضي، بواسطة الأمتصاص الذري للأطياف. أظهرت النتائج وجود إختلافات كبيرة في تركيز المعادن في العينات المختبرة. إذ وجد أن أعلى مستوى لهذه المعادن يمكن أن يرتب أستناداً الى أجزاء النبات المستخدمة الى الحديد في الأوراق، و الزنك في الرايزومات، و النحاس في البذور، و النيكل في البراعم، و الرصاص في الثمار، و الكادميوم في القلف. ففي المعادن الثقيلة أحتوت بذور الجلجلان أو الكراوية على أعلى تركيز من الرصاص (2.2 µg/g) في حين أن تركيزالكادميوم كان الأعلى في بذور الهيل الأخضر (2.9 µg/g). أما بالنسبة للمعادن الأساسية فقد وجد أن أعلى تركيز سجل لكل من الزنك، في حبة البركة (52.2 µg/g) ، و النحاس في الفلفل الأسود (17.3 µg/g) ، والحديد في بذور السنوت (13.6 µg/g) وأخيراً النيكل في حبة البركة (4.9 µg/g) تم تأكيد دقة النتائج والنهج المستخدم بواسطة تحليل البيانات ومقارنتها بما هو موجود في الأدبيات العلمية.

كلمات مدخلية: البحرين، تراكم حيوي، معادن ثقيلة، معادن سامه، بهارات.

Introduction

Spices are dried parts of cultivated aromatic plants, best grown in tropical and subtropical regions of the world. Various kinds have been used for thousands of years by mankind to enhance food flavor, aroma and palatability. Contributing parts of these plants include seeds, bark, roots, leaves, rhizomes and flowers, all of which, when used in small amounts, improve acceptability of foods due to the presence of volatile and fixed oils (Goyer, 1995). Some types of spices, such as caraway, anise, coriander and black cumin, are commonly recognized as condiments or employed for other purposes on account of their fragrance, medicinal values and preservation qualities. Extensive research, oriented toward the commercialization of spices, has been aimed at further exploration of their antioxygenic and antioxidant properties, antimicrobial and antitumor activity, perfumery and cosmetic applications, preservatory actions and food coloring (Sovljanski *et al.* 1990). The main constituents of the dry matter of these plant products are carbohydrates, other organic compounds and to a lesser extent crude fibers and fatty acids (Mecance *et al.* 1960; Al-Kathiri and Al-Attar, 1997). Minerals and other inorganic elements are found in low to

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moderate concentration in these food materials, depending upon the parts used and their place of origin.

Toxic metals are often known as heavy metals or non essential elements (i.e. Pb, Hg, Al, Sn, Ag, Se), which unlike essential elements (i.e. Zn, Cu, Fe, Cl, Mn, Ca), when inhaled or ingested in liquid and food form at high concentration can be harmful. In recent years, much attention has been focused on monitoring the bioaccumulation of toxic metals in medicinal plants and spices (Abdus Sattar and Durrani, 1989), either due to their role in interfering with the vital biological processes or their direct potential toxicity on living biota (Goyer, 1995; Khurshid and Qureshi, 1984). Consumption of heavy metal contaminated plants may impose serious health hazards, such as kidney dysfunction, renal failure, liver damage, chronic symptoms and hypersensitivity symptoms (Friberg *et al.* 1986). Studies confirm that the above metals at high dose can impair neurological function, delay energy production pathways, increase allergic reactions and lower immune system response (Khurshid and Qureshi, 1984).

The sources of environmental pollution with toxic metals are quite diverse, ranging from industrial and traffic emission to the intensive use of broad range human-made chemicals on cultivated crops for high yield production. Abou-Arab *et al.* (1999) proposed two major reasons to evaluate levels of toxic metals in medicinal plants. The first is the increase in the global contamination of the environment with heavy metals. Secondly, herbal remedies, especially those of Asian origin, have repeatedly been reported to have a high concentration of heavy metals. Spices are not unique among other plants which are continually exposed to a polluted atmosphere from industrial wastes, sewage disposals, contaminated irrigation water and heavily loaded soils with toxic chemicals; i.e., fumigants, pesticides, herbicides and fungicides. Toxic metals are of concern in soils affected by heavy concentration of acid rains or close to industrial plants. The increase in acidity stimulates higher solubility of metals as a result, absorbed by plants through roots and thus becoming systemically distributed (Al-Saleh and Chudasama, 1994). Metal can also be deposited directly on plant surfaces (Yoo-Sung and Song-Kung-sik, 1991).

Although levels of selected metals in many biological materials have been established (Janger and Westerlund, 1980; Jelinek, 1982; Benzo *et al.* 1986; Goyer, 1995), their concentration in spices, widely consumed in Bahrain, is not documented. This investigation was planned to evaluate the accumulation of some essential and toxic metals in commonly used imported spices to assure safety and monitor quality for Bahraini consumers.

Materials and Methods

1- Spice collection and preparation

Spice and condiment samples imported from various parts of the world, mainly India, United States and Iran, were collected during the months of March and April 2000. The samples were usually found kept in metal containers, plastic or sometimes glass jars, wooden boxes, or gunny bags and most commonly exposed to air. The materials were obtained from different retail agencies in the spice market (bazaar) of Bahrain in clean, labeled paper bags. Care was taken to avoid old stocks and heavily distorted and contaminated spice samples. In the laboratory, the material was further sorted out based on purity and country of origin, rinsed twice in distilled water and dried in an oven at 60°C on clean sterilized paper towels. The well-dried samples were finely ground in a house blender, sieved through No. 50 mm mesh, kept tightly packed in paper bags and stored at 5°C for further analysis. Every spice sample was analyzed in triplicate. A list of spices, collection code and plant parts used is summarized in Table 1.

2- Moisture content and pH

Thourougly mixed samples were weighed for chemical and physical determination. Ground spice samples were dried at 100°C for 24 hours or until their weight remained constant. The difference in weight represented the moisture content. A 1:10 suspension (spice: distilled water) of each spice was prepared and stirred for 24 hours. The pH of the suspension was measured using JENWAY water analyzer (Model PW1).

Table 1. Common, scientific, and family names and parts used.

Serial code	Collection number	English name	Plant taxon	Family	Plant part used
1	QM 317	Galangale	*Alpinia galanga*	Zingiberaceae	Rhizomes
2	QM 209	Green cardamom	*Ellettaria cardamomum*	Zingiberaceae	Seeds
3	QM 311	Black cardamom	*Amomum angustifdium*	Zingiberaceae	Seeds
4	QM 212	Black pepper	*Piper nigrum*	Piperaceae	Buds
5	QM 221	Red chili	*Capsicum spp*	Solanaceae	Fruits
6	QM 315	Lemon, Dry	*Citrus spp*	Rutaceae	Fruits
7	QM 222	Turmeric	*Curcuma domistica*	Zingiberaceae	Rhizomes
8	QM 310	Ginger, Dry	*Zingiber officinale*	Zingiberaceae	Rhizomes
9	QM 300	Cinnamon	*Cinnamomum zeylanicum*	Louraceae	Bark
10	QM 210	Cloves	*Syzygium aromaticum*	Myrtaceae	Flower buds
11	QM 322	Fennel	*Foeniculum vulgare*	Umbelliferae	Seeds
12	QM 211	Nutmeg	*Myristica fragrans*	Myristicaceae	Peeled seeds
13	QM 333	Coriander	*Coriandrum sativum*	Umbelliferae	Seeds
14	QM 319	Caraway	*Carum carvi*	Umbelliferae	Seeds
15	QM 307	Bay leaf	*Pimenta acris*	Myrtaceae	Leaves
16	QM 293	Cumin	*Cuminum cyminum*	Umbelliferae	Seeds
17	QM 202	Black cumin	*Nigella sativa*	Ranunculaceae	Seeds

3- Chemical analysis

Unless otherwise stated, all laboratory glassware was prewashed with double distilled water and plotted dry before use. Following the procedure of Abou-Arab *et al.* (1999), duplicate samples (0.5 grams) of each spice were weighed in 50 ml Erlenmeyer flasks. For digestion 6 ml of nitric acid (Analar grade) was added to each sample followed by 2 ml of hydrogen peroxide (30%, Puriss grade). The mixture was heated gently on a hotplate for 2 hours or until the volume of the liquid was about 3-5 ml. The material was allowed to cool to room temperature and filtered directly into 50 ml volumetric flasks using Whatman filter paper No. 1. The filtrate was diluted in 50 ml using double distilled water and stored at 5°C. The concentration of metals in the final digest solution was evaluated by means of atomic absorption spectrophotometer (Unicam Atomic Absorbtion). The following metals were determined at wavelength (nm) shown in parentheses Fe (248.3), Zn (213.9), Cd (228.8), Pb (217.0), Ni (232.0), Cu (324.8)nm. The instrument settings were in accordance with the manufacturer's specifications. A series of calibration curves for the metals were prepared by diluting the standard solution (1000 ppm, ISS grade) with deionized water. Deionised water was used as blank solution and a blank reading was taken after every eight sample-readings. Two readings were taken for each sample to insure accuracy of the instrument.

Results and Discussion

Table 2 shows the mean level value of moisture content (%), hydrogen ion concentration (pH) and concentration of some toxic and essential metals in various imported spices. All the results presented in this work were averaged from data derived from at least two replicated procedures. The accuracy and consistency of the obtained values were repeatedly compared with calibration curves of deionized water and was in the order of magnitude of $\leq$ 1% and presented as µg/g.

Moisture levels in the examined spice samples were somewhat high, ranging from 9 to 17.5%, in black cumin and cinnamon, respectively Most of the samples had water content between 10—13%. Galangale, green cardamom and fennel also had somewhat high moisture levels. It is generally observed that seeds, rhizomes, and leaf parts contain higher moisture content than other plant parts. Moreover, water content values were also high when compared with studies conducted elsewhere on spices (Abdus Sattar and Durrani, 1989), probably due to high atmospheric relative humidity in the island of Bahrain (average 60-80%). A substantial increase in relative humidity stimulates faster mold growth on spices that results in rapid quality deterioration and a low marketable value.

Hydrogen ion concentration (pH) was mostly found acidic (average 5.6) (See,table 2). The lowest mean pH of aqueous extracts of spices was reported in dry lemon (2.2) and the highest in dry ginger (7.1). In this study, no clear pattern can be observed in the effect of pH on metal accumulation in plants.

Table 2: Metal concentrations (µg/g), pH and moisture content in spices

Spice code	Spice name	Fe	Zn	Cu	Ni	Cd	Pb	pH	Moisture
1	Galangale	96.8 ±1	24.0 ±2	4.8 ±0.6	2.5 ±0.3	0.07 ±0.0	0.8 ±0.2	5.5	16.4
2	Green Cardamom	61.6 ±7	49.2 ±4	8.1 ±0.7	2.2 ±0.3	0.90 ±0.1	0.7 ±0.1	6.5	14.2
3	Black cardamom	83.7 ±5	33.4 ±3	8.7 ±0.4	1.6 ±0.2	0.04 ±0.0	1.0 ±0.3	5.0	12.2
4	Black Pepper	56.5 ±0.3	12.4 ±1	17.3 ±0.3	4.3 ±0.2	0.02 ±0.0	0.4 ±0.05	6.1	9.8
5	Red Chili	52.5 ±6	20.0 ±0.5	9.2 ±0.4	2.2 ±0.2	0.15 ±0.05	0.9 ±0.1	4.7	13.7
6	Dry Lemon	58.2 ±7	9.2 ±3	6.6 ±0.2	1.8 ±0.1	0.01 ± 0.00	1.2 ±0.2	2.2	13.5
7	Turmeric	110.8 ±7	9.8 ±0.5	7.2 ±0.5	2.4 ±0.6	0.03 ±0.01	0.8 ±0.06	6.5	12.7
8	Dry Ginger	104.7 ±33	13.7 ±2	5.2 ±0.2	1.8 ±0.2	0.05 ±0.02	0.5 ±0.1	7.1	13.0
9	Cinnamon	49.7 ±6	12.7 ±1	6.9 ±0.4	2.5 ±0.2	0.25 ±0.02	1.6 ±0.2	4.9	17.5
10	Cloves	63.2 ±11	12.3 ±0.8	6.0 ± 0.5	2.1 ±0.1	0.07 ±0.04	0.7 ±0.05	6.0	9.3
11	Fennel	116.9 ±11	26.4 ±0.3	11.8 ±0.2	3.1 ±0.4	0.10 ±0.01	0.6 ±0.06	5.8	14.8
12	Nutmeg	37.3 ±2	16.5 ±0.5	15.8 ±5	2.6 ±0.1	0.06 ±0.05	0.7 ±0.05	6.2	10.4
13	Coriander	40.8 ±0	36.0 ±0	11.5 ±0	3.0 ±0	0.19 ±0	1.1 ±0	6.7	12.8
14	Caraway	96.3 ±0	37.1 ±0	14.9 ±0	1.8 ±0	0.17 ±0	2.2 ±0	6.7	11.9
15	Bay leaf	113.0 ±9	15.6 ±0.9	9.1 ±0.2	3.5 ±0.3	0.04 ±0.00	0.9 ±0.1	5.3	13.0
16	Cumin	133.6 ±0	34.4 ±0	8.5 ±0	3.1 ±0	0.03 ±0	1.0 ±0	6.0	10.9
17	Black Cumin	123.8 ±0	52.2 ±0	13.5 ±0	4.9 ±0	0.06 ±0	0.6 ±0	5.9	9.0

Figures 1 and 2 show the overall distribution pattern of the six evaluated metals among the various spices. In general, heavy metal (Cd, Pb) concentrations were observed far below (range 0.01—2.2 µg/g) those of essential elements (Fe, Zn, Cu, Ni) (range 1.6—133.6 µg/g). Concentration across the different plant parts revealed that the maximum accumulation levels of essential metals were noted in leaves, rhizomes, seeds, buds, fruits, and barks in the order of magnitude of Fe>Zn>Cu>Ni, respectively. Heavy metal, on the contrary, showed an irregular distribution pattern among plant parts. Cadmium contamination in spices was considerably low and fluctuated from 0.01mg/g in dry lemon to 0.9 µg/g in green cardamom and consequently followed by cinnamon (0.25 µg/g) (Figure 1). Clearly, seeds and bark contained more cadmium than other plant parts (See,table 2). Among the most important foods possibly contaminated with cadmium are pork, fish, milk and beer. In addition, cadmium becomes more soluble in acid foods and drinks like fruit juices and vinegar. Goyer (1995) reported that a normal diet supplies approximately ≤ 0.01-0.04 µg/g of cadmium per day, depending on the consumed amount of food.

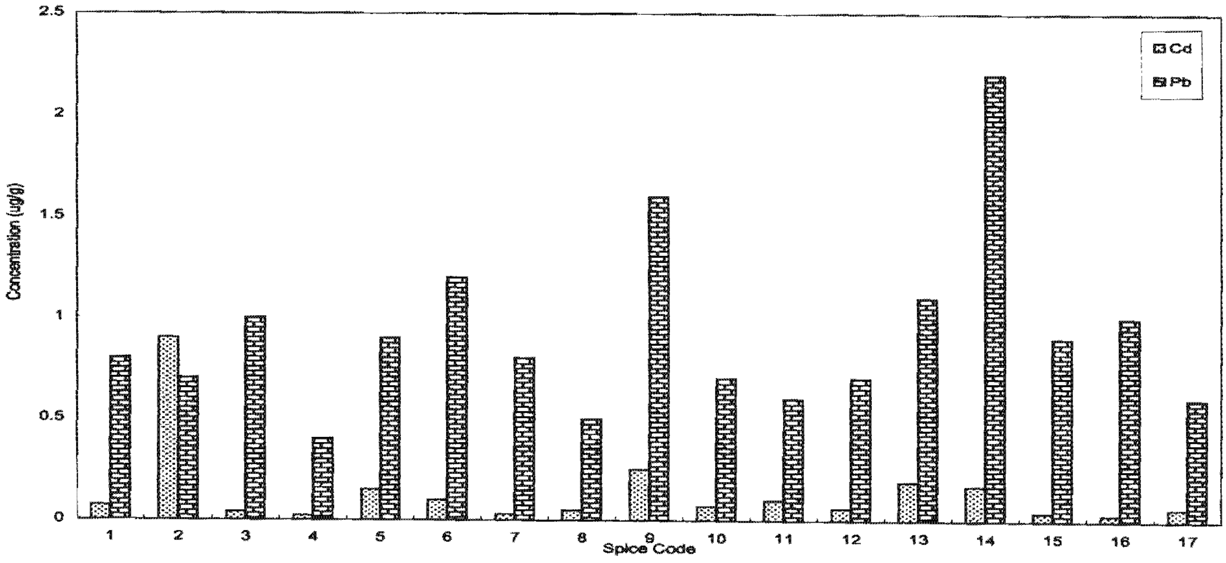

Figure 1.: Concentrations (µg/g) of heavy metals in commonaly used spices in Bahrain

Lead concentration in the tested samples varied from 0.4 to (2.2 µg/g) with a mean average of (0.9 µg/g.) Maximum lead levels were detected in caraway (10 µg/g) followed in descending order by cinnamon (1.6 µg/g) and dry lemon (1.2 µg/g). Coriander, cumin, black cardamom and bay leaf also had considerable amounts of lead (See,figure 1). Overall mean level of lead accumulated across all spice samples was found considerably higher (range 0.4—2.2 µg/g) than the cadmium level (range 0.01—0.9 µg/g).

Recently, Abou-Arab *et al.* (1999) attributed high concentration of lead and cadmium in different medicinal plants to use of contaminated irrigation water as well as the addition of some chemical fertilizers and herbicides. In many industrialized countries, average lead intake from food and beverages has been estimated for adults to be (25-30 µg/g) per day (Gravel *et al.* 1994; Januz *et al.* 1994).

Our findings on lead and cadmium concentration were comparable with those of spices reported by Abdus Sattar and Durrani (1989) from Pakistan, Listow and Petrow (1990) from USSR, Bosque *et al.* (1990) from Spain and, for lead only, medicinal plants from Egypt (Abou-Arab *et al.* 1999), while cadmium levels were much lower. On the other hand, levels of both heavy and essential metals are much lower than those obtained by Jawad *et al.* (1986), Al-Saleh and Chudasama (1994) and Al-Kathiri and Al-Attar (1997) from Iraq, Bahrain and Saudi Arabia, respectively. Friberg *et al.* (1986) reported that lead accumulation in food varied from 0.01 to 2.5 µg/g, whereas the narrower cadmium range varied from 0.005-6.1 µg/g.

Several reports in the literature have indicated that accumulation of heavy metals is a result of many factors, e.g. traffic emissions, farm location and distance from roadsides (Marletta *et al.* 1986; Bosque *et al.* 1990). Lead is known to deposit on biological material as a consequence of aerosol fine particles and vapor, even at a remote distance from traffic or industry emission, while pollution with cadmium is more pronounced only for leafy crops grown at locations adjacent to highways. Gravel *et al.* (1994) and Januz *et al.* (1994) compared several locations for deposition of heavy metals in medicinal plants. They concluded that plants grown in regions of dense industry accumulate higher levels of heavy metals than plants grown in less industrialized regions due to greater contamination levels of air and soil.

Among various organically grown vegetables and field crops in different major US agriculture areas, Wolnik *et al.* (1985) showed that mean concentrations of lead and cadmium in tomatoes and onions were 0.17 and 0.11mg/g, respectively. In addition, Kim *et al.* (1994) estimated the content of metals in 291 samples of medicinal plants grown in rural unpolluted sites in Korea. They concluded that concentrations of Cd, Cu, Pb, Zn, Cr, Ni and As in these plants were 0.38, 6.6, 0.81, 27.7, 1.45, 0.73 and 0.27 µg/kg, respectively. These results support our findings for the concentrations of heavy metals in spices; moreover, overall cadmium concentration was considerably lower in all the examined samples.

The maximum mean concentration of copper was found in black pepper (17.3 µg/g), followed by nutmeg (15.8 µg/g), caraway (14.9 µg/g), black cumin (13.5 µg/g), fennel (11.8 µg/g) and coriander (11.8 µmg/g) (Fig. 2).

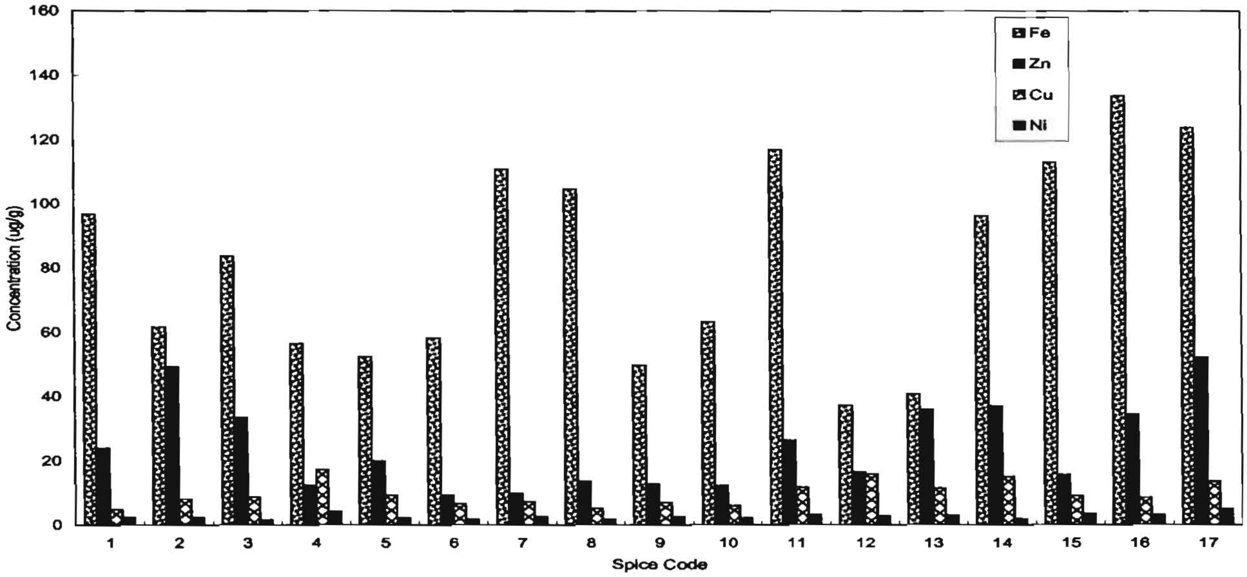

Figure 2.: Concentrations (µg/g) of essential metals in commonaly used spices in Bahrain

Other remaining samples contained a varied range (4-9 μg/g) of copper. The minimum mean level of this metal was noted in galangale (4.8 μg/g). Copper is an essential element when consumed ≤10 mg/kg and involved in redox reactions; however, a serious health hazard can result when a formula of copper sulfate is used as fungicide.

The mean level of nickel varied considerably among spice materials, fluctuating between 1.6mg/g in black cardamom and 4.9 μg/g in black cumin (See, fig. 2). The highest mean concentration of nickel was observed in decreasing order pattern at 4.3, 3.5, 3.1 and 3.1mg/g in black pepper, bay leaf, fennel and cumin, respectively. All other samples had cobalt content ≤ 3mg/g.

Spice samples analyzed for zinc reveal mean levels of as high as 52.2 μg/g to as low as 9.2 μg/g. The maximum mean concentration of zinc was detected in black cumin, followed in a decreasing order by green cardamom (49.2 μg/g), caraway (37.1 μg/g), coriander (36 μg/g), cumin (34.4 μg/g) and black cardamom (33.4 μg/g). The overall mean occurrence of zinc in all spice samples is 24.4mg/g (See, fig. 2).

Iron content was the highest among all the examined spice samples and was not found below 37 μg/g. Mean level content varied between 37.3 – 133.6 μg/g for nutmeg and cumin, respectively. Black cumin, bay leaf, fennel, turmeric, dry ginger, caraway and galangale also had considerable amounts of iron >90 μg/g.

Table 3 compares the highest and lowest levels of heavy and essential elements in spice samples. It is obvious that the decreasing order pattern of these metals are Fe > Zn >Cu>Ni>Pb>Cd in the following concentration average of 82.3, 24.4, 9.7, 2.7, 0.9 and 0.13 μg/g, respectively. Nevertheless, considerable variations were noted among the various spices.

Table 3: Highest and lowest concentration of essential and toxic metals in widely used spices in Bahrain.

Element	Symbol	Highest levelmg/g	Lowest level mg/g	Mean mg/g
Iron	Fe	133.6	37.3	82.3
Zinc	Zn	52.2	9.2	24.4
Copper	Cu	17.3	4.8	9.7
Nickel	Ni	4.9	1.6	2.7
Cadmium	Cd	0.9	0.01	0.13
Lead	Pb	2.2	0.4	0.9

Shroeder (1971) reported that various spices generally contained on the average higher zinc levels (23 μg/g) than copper (6.8 μg/g). The data for essential trace metals were also comparable to those reported by other workers (Mecance *et al.* 1960; Benzo *et al.* 1986). Recently, Abou–Arab *et al.* (1999) analyzed various spices and medicinal plants for metal content and showed that biological samples contained on average ≥ 72.44 μg/kg iron, 64.2 μg/g zinc, 33.8 μg/g manganese, 5.5 μg/g copper and 0.053 μg/g cobalt. These findings support our results in most cases for essential trace metals and also coincide with those of Abdus Sattar and Durrani (1989) on spice samples of Pakistan.

Since heavy metals (Pb and Cd), are not essential for any metabolic processes in the cell, they are considered to be toxic pollutants in food and their presence should be of immediate concern (Wolnik *et al.* 1985).

The results, as a whole, indicate that spices examined in the current work vary widely in their toxic and essential metal contents. Concentration of toxic metals in spice samples were lower than other reports, while levels of Fe, Zn, Cu and Ni were almost comparable, due to, in part, location relative to traffic and industry emission, agricultural practices, plant spices and climatic factors (Bosque *et al.* 1990). Guideline data for heavy metals are not available in developing countries. Background information is needed for evaluating the toxicological and nutritional significance of these elements in spices and their possible increase due to various pollution hazards. Therefore, it is of immediate concern to monitor element contents on a continuous basis in spices and any other biological materials.

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