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ASSESSMENT OF HEAVY METAL CONTENTS IN SOME COMMON SPICES AVAILABLE IN THE LOCAL MARKETS OF TIKRIT

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ABSTRACT

The existence of the heavy metals in spices, which is primarily used for coloring and flavoring food, has serious threats on human health owing to their high toxicity at high concentrations. This work deals with estimating the contents of copper (Cu), cadmium (Cd), iron (Fe), nickel (Ni) and lead (Pb) in ten different types of common spices available in the local markets of Tikrit. Five types of spices were purchased packed in tightly closed boxes, which are (biryani spices, turmeric, dolma, fish, mixed spices) while the others were purchased uncovered in wooden boxes or bags inside or outside the stores, which are (chicken shawarma spices, kebbeh, burker meat, curry, yellow meat broth). After the wet digestion process of each sample was conducted, the concentrations of the heavy metals based on dry weight (ppm) were estimated using the atomic absorption spectrophotometer. The digestion process and estimation of heavy metal contents were conducted in three replicates for each spice. The results demonstrated that the concentrations of heavy metals in the spices ranged between 0.584 - 3.814 ppm for copper, 0.074 - 0.885 ppm for cadmium, 6.870 - 19.370 ppm for iron. 0.521 - 1.356 ppm for nickel, and 0.014 - 0.161 ppm for lead. The copper, cadmium, iron, nickel, and lead metals were largest in the turmeric, kebbeh, curry, mixed, and curry spices respectively. These concentrations were compared with the results available in literature, organizations, and associations to determine whether these concentrations are appropriate and safe. The results showed that most of the concentrations of heavy metals in the spices studied herein were within the permissible limits and safe for human consumption.

KEY WORDS : Heavy metals, Spices, Food poisoning, Atomic absorption spectrophotometer

INTRODUCTION

Recently, herbs, medicinal plants, spices, and perfumes occupy an important place in most countries around the world. Spice is one of the important groups in the agricultural trade that is indispensable in the kitchen, as they are used for flavoring and coloring food. Spices are aperitive and can increase the liquor gastricus owing to their high values, flavors, colors, tastes, and distinctive aromas. They are used whole, ground or extracts and added during the preparation and processing of various foods (Arafa, 2004). Spices are defined according to the International Standards Organization (ISO) as natural plant products that are used whole or ground. Both groups of "herbs and spices" are distinguished by their pungent odor and taste, and some of them have distinctive colors. In addition to the usage of spices and herbs for flavoring and coloring food, they are used for their medicinal benefits. Al-Adaida (Abdul Majeed, 2007). They are also used as preservatives as well as their aromatic characteristics have a benefit in removing undesirable scent in some foods. Spice plants often descend in their origin to the Mediterranean region, and tropical spices are often characterized by their spicy like pepper or their high aromatic content (Al-Hilali *et al.*, 2021). The European Spice Association

defined cooking herbs and spices as parts taken from different places of plants or dried herbs used for cooking, and they are the edible parts of plants (Kaprinska *et al.,* 2001).

Spices are same as most plants in exposure to a wide range of microbial contaminants before, during, and/or after collection and/or during commercial drying processes. Contamination also occurs during manufacturing, importing, exporting, storage, promotion, sale and/ or usage (Ramesh, and Jayagoudar, 2013). Most of the common spices have been documented to have microbial potential, and when prepared and dealt with, they can make them a source of food poisoning. Moreover, spices may become contaminated with heavy metals which comes from several sources including the atmosphere and industrial atmospheric deposits represented by the chemical industries as their wastes, either in the liquid form that is discharged into seas and rivers, or in the gas form, contain high percentages of these metals (Mubeen *et al.,* 2009). Contamination of spices by minerals can also be attributed to the agricultural practices such as fertilization, spraying of pesticides as well as irrigation by wastewater contaminated with heavy metals. Farming in contaminated soil and packing materials are another source of pollution (Hinne Bur Gi *et al*., 2006).

Mammals, including humans, are affected by heavy metals. The effect of the heavy metals on human health depends on their percentage, toxicity, and how the human exposed to them. They are considered dangerous because they tend to accumulate inside the body and their concentration increases with time in comparison to their chemical concentrations in the nature (Srini-Vasan *et al*., 2005). Spices are a potential link for transporting pollutants to humans through the food chain due to their plant origin (Soliman *et al*., 2015). The addition of spices contaminated with heavy metals to food may lead to the accumulation of these metals in the organs of the human body and may affect its health in terms of the bioaccumulation potential as these metals are toxic to living organisms even in low concentrations and are not decomposed in their bodies. Consequently, the monitoring of heavy metals in crops and other foods is of great importance in protecting humans from the hazards of these metal ions (Abou-Arab *et al.,* 2000; Ba°gel *et al*., 2006). The exposure to toxic metals is associated with serious health problems including kidney problems, nervous behavior, growth problems and disorders, high blood pressure and possibly cancer (Maiga *et al.,* 2005; Galanis *et al*., 2009).

Owing to the importance of this topic and its solid relation to human health and the nature of his food, a survey study on the contamination of the most used spices in our society by heavy metals was conducted to examine their extent of contamination and to focus light on the spices that are most vulnerable to the contamination by the heavy metals, which are considered among the most toxic pollutants and dangerous to human health if they increase than the permissible level.

MATERIALS AND METHODS

Materials

The spices used in this work were purchased from the local markets of Tikrit. Five of them were purchased packed in tightly closed boxes, which are (biryani spices, turmeric, dolma, fish, mixed spices) while the others were purchased uncovered in wooden boxes or bags inside or outside the stores, which are (chicken shawarma spices, kebbeh, burker meat, curry, yellow meat broth). Three samples for each spice studied herein were purchased from different places within the city and placed in polyethylene bags, then kept in the refrigerator at 4 °C until they were used.

Preparing work tools

All tools and glass ware used in this work were washed thoroughly with distilled water, then immersed in concentrated nitric acid solution, then washed again with distilled water for several times, and then the samples were digested.

Work methods

The spices samples were dried using an oven at a temperature of 80 °C for 12 hours in order to obtain completely dry samples. After that, the samples were crushed in a ceramic mortar and sieved through a sieve with pore size of 0.5 mm. 0.5 g of each sample was then placed in a ceramic crucible and the digestion process was carried out by the wet method mentioned in literature (Jones *et al.,* 1990; Kalra, 1998). The digestion process was performed by adding 3.5 ml of solution consisting of 30% of hydrogen peroxide H_2O_2 and 2 ml of sulfuric acid H_2SO_4. The mixture was heated to a temperature of 100 °C, and then the temperature was raised to 250 °C. The heating process lasted for 30 minutes, then the mixture was left to cool down. After that, 1 ml of

30% H_2O_2 was added to the mixture and the heating process was repeated until the bright color of the digestion product was attained. The mixture was then cooled and filtered using a filter paper in a 25 ml glass beaker and the volume was completed to 25 ml by adding deionized water.

Measurement of heavy metal ions

The heavy metals studied in this work were copper Cu, cadmium Cd, iron Fe, nickel Ni, and lead Pb. The standard solutions for the tested metals were prepared according to the methods mentioned in literature (Inam *et al.*, 2013). The heavy metals were estimated in each sample using the atomic absorption spectrophotometer (model Shimadzu 7000-AA).

RESULTS AND DISCUSSION

The presence of heavy metals in nature and in the food of humans and animals within the permissible limits is acceptable; however, an increase in their concentrations beyond the permissible limits is undesirable due to the presence of the bioaccumulation characteristic of these heavy metals. This study included the measurement of the heavy metal contents in different types of spices available in the local markets of Tikrit. Significant differences were found between the contents of heavy metals in the samples studied herein in comparison to the maximum permissible limits for heavy metal contents in spices according to the World Health Organization and the Food and Agriculture Organization (FAO/WHO2010). Table (1) shows the permissible limits for heavy metals in spices according to the organizations mentioned early.

Samples' Content of Copper

The increase of the copper concentration in the human body beyond a certain limit causes several health problems, the most important are high blood pressure, anemia, and nervous system disorders. It also leads to behavioral disorders in children, dermatitis, liver damage and can cause cancer (Onianwa *et al.*, 2001). Figure 1 shows the copper concentration in the spices studied in this work. The outcomes demonstrated that the concentration of the copper element in the studied spices ranged between (0.584 - 3.814) ppm. The lowest concentration value was noticed in the mixed type spices, while the highest concentration value was noticed in the turmeric spices. The results also showed that the copper contents in all samples were within the limits permitted by the WHO / FAO which is 30 ppm. In comparison to the results obtained from other studies, it was noticed that the concentration of copper herein was lower than that mentioned in a study which was conducted to estimate the concentration of copper in some common spices in the markets of Basra, which ranged between 5.20 and 8.35 ppm (Mustafa *et al.*, 2020). Another study conducted in Baghdad reported that the copper concentration reached its highest concentration in black pepper with a value of 0.6525 ppm, while the its lowest concentration was in turmeric and was of 0.5244 ppm (Al-Timimi and Jaber, 2015). The concentrations of copper in all studied samples herein were within the permissible limits according to the International Food and Agriculture Organization and therefore this heavy metal has no toxic effect and does not cause any harm to the consumer's health in the long term.

Table 1. Permissible limits for heavy metals in spices, estimated in ppm according to WHO / FAO

The studied metals	Permissible concentrations for heavy metals in spices (ppm)
Copper	30
Cadmium	0.2
Iron	300
Nickel	1.63
Lead	5

Samples' Content of Cadmium

The exposure of human to this metal, even in small proportions for long time, leads to kidney problems, lung diseases, osteoporosis, high blood pressure,

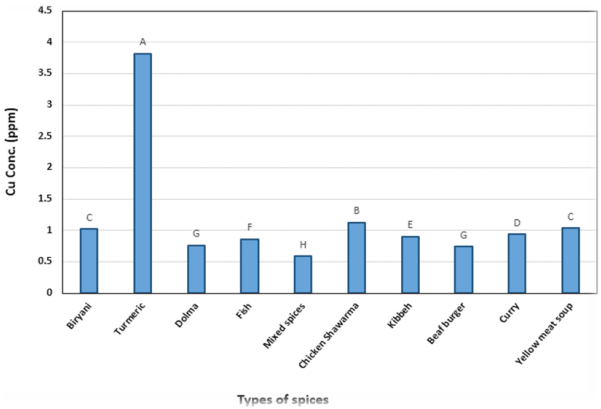

Fig. 1. Copper concentration in ppm for the studied spices.

problems in liver function, and the destruction of brain cells. Additionally, it works to break down fatty acids, and is considered carcinogen and the increase in its concentration in food causes harm to the health of the consumer (Ozkutlu *et al*., 2006; Arpadjan, *et al*., 2008). Figure (2) illustrates the cadmium concentration in the spices studied in this work. The outcomes showed that the concentration of the cadmium element in the studied spices ranged between (0.074 -0.885) ppm. The lowest concentration value was noticed in the curry type spices, while the highest concentration value was noticed in the kibbeh and Berker meat spices. These concentrations were high in comparison to the global permissible limits according to the World Health Organization which is 0.22 ppm. The cadmium concentrations in the current study was lower than these reported, as they indicated that the concentration of cadmium in some types of spices in the markets of Basra city ranged between 5.6 - 8.1 ppm. They also reported that their results did not agree with the outcomes from another study (Al-Timimi and Jaber, 2015). Other similar studies including the one conducted in the city of Baghdad reported that the cadmium concentration ranged between 0.0021 - 0.1975 ppm in some types of spices. The obtained results in this work were not also in agreement with the study made by some researchers to estimate the heavy metals of spices common in the Ghanaian market. They found that the cadmium concentration was higher than the concentration of other metals and had the range of 0.02 - 0.58 ppm (Crentsil *et al*., 2012). The reason behind the high concentration of this metal is probably due to the contamination of the site's soil used in agriculture. Whereas, the variation among results may be due to the difference in distances between the farming site and the pollution source in addition to the quality of the irrigation water in which the using of untreated water leads to the accumulation of heavy metals in the spices.

Samples' Content of Iron

Iron is one of the important nutrients for the human body, and the deficiency of its level in the blood causes many health damages, including anemia; however, the increase in iron concentration leads to iron toxicity and may include early symptoms of iron poisoning, stomach pain, nausea, and vomiting, in addition to causing cirrhosis, heart failure, and diabetes (9, 11). Figure 3 illustrates the iron concentration in the spices studied in this work. The results demonstrated that the concentration of the iron element in the studied spices ranged between (6.870 - 19.370) ppm. The lowest concentration value was noticed in the dolma type spices, while the highest concentration value was noticed in the curry spices. All the results were within the limits permitted by FAO/WHO which is 300 ppm. When comparing the results of this study, it can be seen that they were in agreement with the results of the study conducted on the spices in the Baghdad city which reported that the iron concentration ranged between (3.48-81.35) ppm (20). However, the

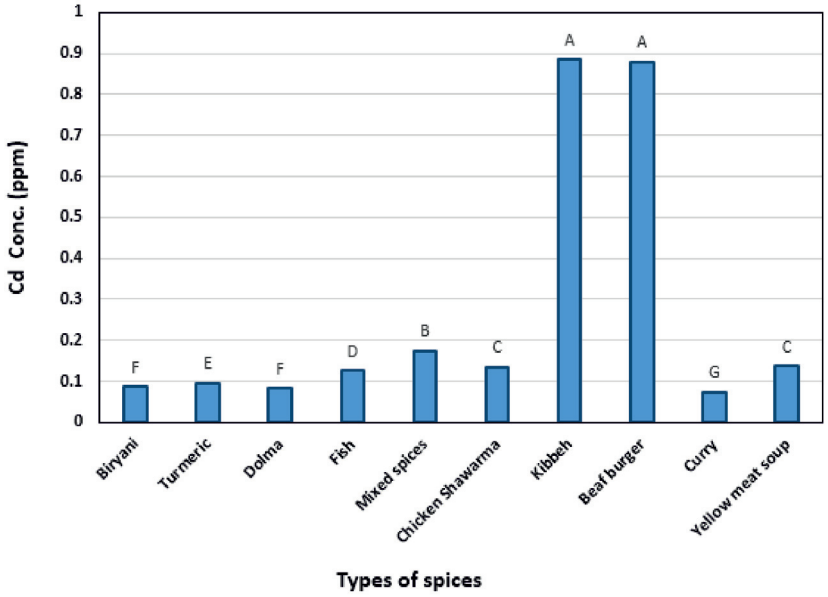

Fig. 2. Cadmium concentration in ppm for the studied spices.

concentration of iron metals in this study was lower than that from the study conducted to estimate the iron concentration in some common spices in the markets of Basra city. The latter reported high iron concentration ranging between 79.15-186.60 ppm. The difference in the iron concentration is likely due to the wide spread of iron in the soil as well as air pollution with this metal which varying from place to another. It is also likely dueto the absorption of the iron metal during the grinding process of these spices or because the pesticides used.

Samples' Content of Nickel

Nickel is considered a health hazard if it enters the human body. The increase in its concentration causes damage to kidney tissues, liver disorders, as well as damage to the lung and blood circulation. The International Center for Cancer Research classifies nickel as a cancer-causing substance for human and the continuous exposure to nickel is one of the causes of lung cancer (6) and (17). Figure (4) shows the nickel concentration in the spices studied in this work. The results demonstrated that the concentration of the nickel element in the studied spices ranged between (0.521-1.356) ppm. The lowest concentration value was noticed in the curry spices, while the highest concentration value was noticed in the mixed type spices. These results were within the limits internationally permissible according to FAO / WHO which is 1.63 ppm, and therefore these spicesare not deemed a risk to human health which also proven by as many studies performed to estimate the daily human exposure limit for this metal (Olnsakin and Olaouwa, 2016). When comparing the results of the current study with other studies, it can be said that the concentrations of nickel element hereinwere less than those obtained by a study conducted in Abuja, Nigeria to determine heavy metals in commonly used spices (Umar *et al.*, 2014). The latter study stated that the concentration of nickel element in common spices in Abujaranged between (96.5-19.4) ppm.

Samples' Content of Lead

Lead is a cumulative-toxic substance that affects many human body systems and harms young children in particular due to its high toxicity. Owing to its dangerous influence on the health and safety of living organisms and the ability of these organisms to accumulate and concentrate this element, very small concentrations of it are considered very dangerous (Simonoff *et al*., 1993). The lead metal is considered a direct competitor to calcium inlink to protein uptake sites in the mucous membrane of small intestine. Lead poisoning is concentrated in the bones, blood, kidneys, brain, and thyroid gland and causes deficiencies in its work and leads to mental retardation in children (Ziyaina *et al*., 2014). Figure 5 shows the lead concentration in the spicesstudied in this work the highest concentration value was noticed in the curry spices (0.161 ppm) followed by the meat Berker spices (0.155 pm), while the lowest concentration value was noticed in the mixed type spices (0.0140 ppm). These concentrations were low in comparison to the global

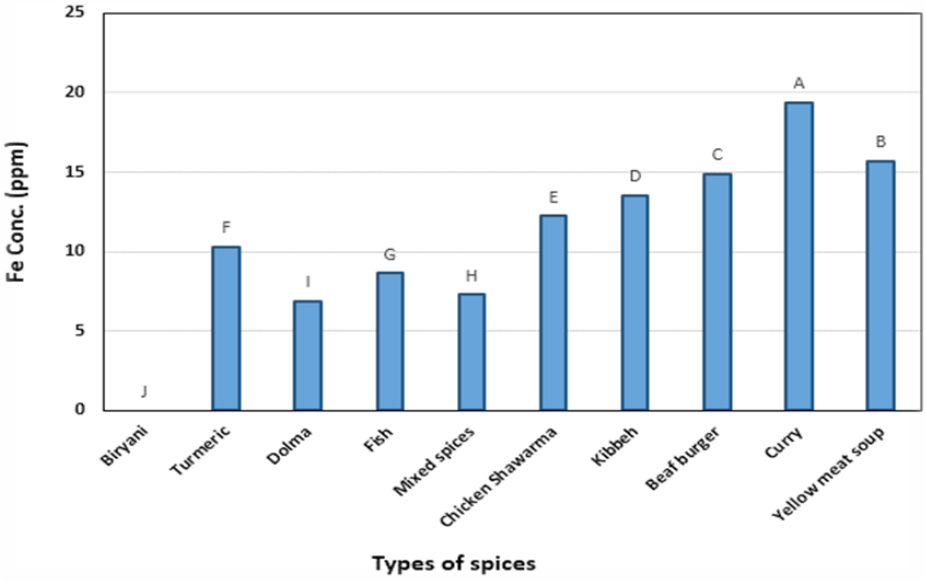

Fig. 3. Iron concentration in ppm for the studied spices.

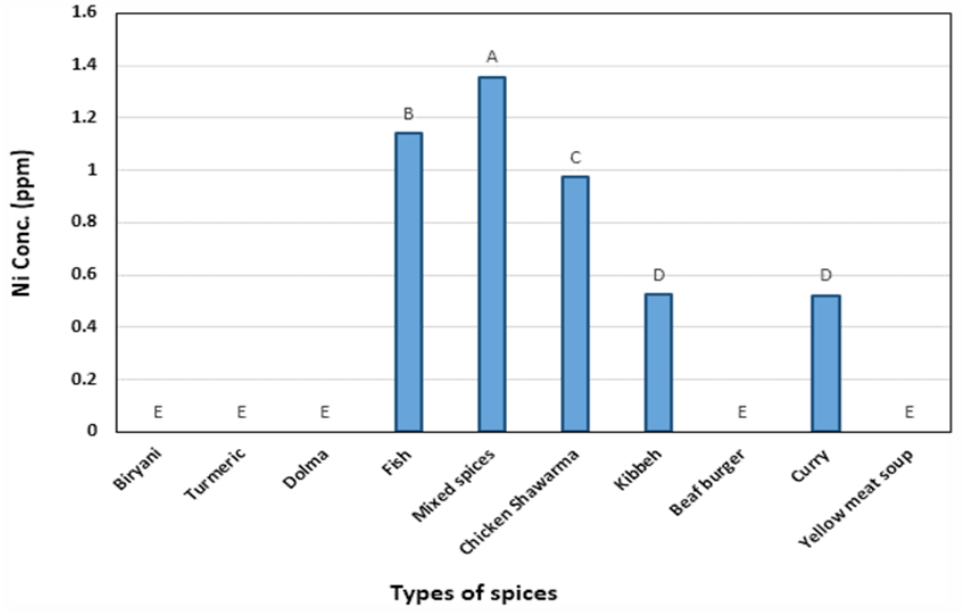

Fig. 4. Nickelconcentration in ppm for the studied spices.

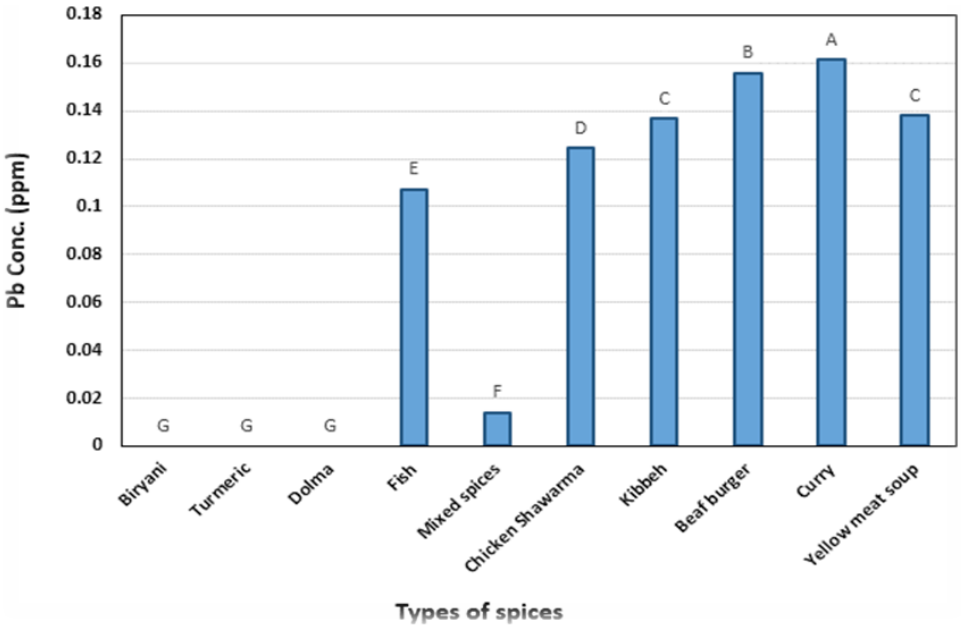

Fig. 5. Lead concentration in ppm for the studied spices.

permissible limits according to the FAO / WHO which is 5 ppm, and consequently these spices are not deemeda risk to human health.

CONCLUSION

It can be concluded that nearly all types of common spices available in the city of Tikrit are not contaminated with heavy metals, apart from kibbeh, Berker meat, and curry spices whichwere characterized by high cadmium in them. Consequently, they should not be overused because it can represent a risk to the health of consumers. It was also found that the concentrations of the other heavy metals in the spices studied in this work were generally low and within the global permissible limits which meet the appropriate safety standards that have no effect on health.

Recommendations

1. Issuing a local standard for spices to be used as a guide in evaluating their quality.

2. Increase the monitoring over imported spices and banning imports from countries whose shipments exceed the permissible limits of the heavy metal concentrations.
3. Paying attention for studying spices and the harmful substances they may containdue to their frequent use in food and the diseases that these harmful substances may cause to consumers.

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