

Determination of Heavy Metal levels in Common Spices

Al-Eed, M. A., F. N. Assubaie, M. M. El-Garawany*, H. EL-Hamshary
and Z. M. ElTayeb

*Dept. of Chemistry & Botany, College of Agricultural & Food Sciences
King Faisal University, P.O. Box 420, Al-Hasa 31982, Saudi Arabia*

ABSTRACT

The concentrations of some heavy metals such as lead (Pb), cadmium (Cd), cobalt (Co), and selenium (Se) present in common spices available at local markets in Saudi Arabia were determined using atomic absorption. The study showed differences in metal concentrations according to the edible part (root, stem, leaf, and fruit). The concentration of lead (Pb) ranged from trace to 14.30 mg kg^{-1} on dry weight basis, where as that of cadmium (Cd) was ranged from 1.25 mg Kg^{-1} to 3.05 mg Kg^{-1} . The concentration level of cobalt was from zero to 0.64 mg kg^{-1} . While variable levels of selenium were detected from zero to 13.3 mg kg^{-1} . Some of these concentrations are above the standard limit approved by WHO and FAO. No risk from daily intake of the most of spices under study for hazardous Pb, Cd, Co, and Se if the human take about 20 g of spices per day. But there are dangrous from basisic, thym and ginger for lead. While the dangrous of cadmium is from fenugreek.

*Permanent address: Soil Fertility and Plant Nutrition Res. Dept., Soil, Water and Environ. Res. Inst., Agric Res. Center. Egypt.

INTRODUCTION

The widespread of contamination with heavy metals in the last decades has raised public and scientific interest due to their dangerous effects on human health (Gilbert 1984). This has led researchers all over the world to study the pollution with heavy metals in air, water, and foods to avoid their harmful effects (Zakrzewski, 1991; Kennish, 1992; Oehme, 1989), and to determine their permissibility for human consumption.

Spices are dried parts of plants, which have been used as diet components often to improve color, aroma, palatability and acceptability of food. They consist of rhizomes, barks, leaves, fruits, seeds, and other parts of the plant (Wahid et al. 1989). Most of these are fragrant, aromatic and pungent. The bulk of the dry material of spices contains carbohydrates, and organic compounds having diverse functional groups. The addition of spices -that may be contaminated with trace and heavy metals- to food as a habit may result in accumulation of these metals in human organs and lead to different health troubles.

Heavy metals are those with atomic weights from 63.546 to 200.590 (Kennish 1992), and specific weight higher than 4 (Connell et al. 1992). These metals may reach and contaminate plants, vegetables, fruits and canned foods through air, water, and soil during cultivation (Husain et al, 1995; Ozores et al, 1997; Geert et al., 1989), and also during industrial processing and packaging (Ibert, 1984; Tsoumbaris et al., 1994).

Thus several studies were done to determine the concentration of heavy metals in spices, dry fruits, and plant nuts (Wahid et. al. 1989, Gilbert 1984, Husain et al, 1995), and to study their dangerous effects. Subjecting to trace and heavy metals above the permissible affect the humman health and may result in illness to human fetus, abortion and preterm labor, and mental retardation to children. Adults also may experience high blood pressure, fatigue and kidney and brain troubles (FAO 1984).

The Kingdome of Saudi Arabia (KSA) imports spices among a lot of food stuff from everal countries. These spices may be subjected to contamination by way or more as described above. We are not aware of published data or results about the contamination and concentration of trace and heavy metals in spices available in the local markets of K. S. A. except that of Selim et. al., 1994, and Al-Eed et. al., 1997, which were done for a very few kinds of spices.

The objective of this work is to estimate the levels of some heavy metals i.e. lead, cadmium, cobalt, and selenium that may be present in spices available in local markets in Al-Hasa region. Also, the levels of investigated metals were recommended by the International Organizations (FAO and WHO).

Material and Methods

Spices Samples were collected from local markets, Recognized and classified according to their English name, scientific name, and the used part of the plant (Table 1). Sample origin is not specified.

Sample preparation

Samples were cleaned and oven-dried at 80 C° for ≈ 12 hrs before chemical analysis. The dried samples were ground in a stainless steel mill till obtaining fine particles that pass through a 0.5 mm mesh and kept dry for analysis.

Table I. Scientific and common names of studied spices

Common name	Scientific name	Family	Used part
Cumin	<i>Cuminum cyminum</i>	Umbelliferae or Apiaceae	Seeds
Coriander	<i>Coriandrum sativum</i>	Umbelliferae or Apiaceae	Seeds
Nigella	<i>Nigella sativa</i>	Umbelliferae or Apiaceae	Seeds
Cinnamon	<i>Cinnamomum zylanicum</i>	Lauraceae	Bark
Fenugreek	<i>Trigonella foenumgraecum</i>	Leguminosae	Seeds
Basilic	<i>Ocimum, ssp</i>	Labiatae	Leaves
Nasturtium	<i>Lepidium sativum</i>	Cruciferae	Seeds
Mahalib	<i>Prunus mahalib</i>	Labiatae	
Turmeric	<i>Cucuma longa</i>	Zingiberaceae	Rhizomes
Thyme	<i>Thymus vulgaris</i>	Labiatae	Leaves
Black pepper	<i>Capsicum nigrum</i>	Piperaceae	Seeds
Ginger	<i>Zingiber officinalis</i>	Zingiberaceae	Rhizomes
Safflower	<i>Cathamus tinctorius</i>	Asteraceae	Petals
Cardamon	<i>Elettaria cardamonum</i>	Zingiberaceae	Seeds

Garden sage	<i>Salvia officinalis</i>	Labiatae	
Nutmeg	<i>Myristica fragrance</i>	Myristicaceae	
Anise	<i>Pimpinilla anisum</i>	Umbellifeae or Apiaceae	
Mahaleb cherry	<i>Prunus mahaleb</i>	Rosaceae	

Determination of metal concentration

For determination of heavy metal concentrations, a wet digestion of the dried samples was done according to the method described by Jones and Case (1990) using conc. H₂SO₄ and 30% H₂O₂ mixture. To a 0.5 g of dry-ground sample placed in 100-ml beaker, was added 3.5 mL of 30 % H₂O₂. The content of the beaker was heated to 100 °C, and the temperature was gradually increased to 250 °C, and left at this temperature for 30 min. The beaker was cooled and more 1 ml of 30 % H₂O₂ was added to the digestion mixture and the contents were reheated again. The digestion process was repeated more than one time until clear solution was obtained. The clear solution was transferred into 50-ml volumetric flask, and completed to the mark with double distilled deionized water. A blank digestion solution was made for comparison. A standard solution for each element under investigation was prepared and used for calibration. Metal measurement was performed with a Perkin-Elmer model 2380 Atomic Absorption Spectrometer, double beam and deuterium background correction. Hollow cathode lamps of Pb, Cd, Co and Se were used at specific wave length of every metal. Measurements were done against metal standard solutions.

The daily intake (mg kg⁻¹ day⁻¹) was calculated based on these suppose

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

$$\text{The daily intake (mg kg}^{-1}\text{ day}^{-1}\text{)} = \text{metal concentration in spice} \times 20/1000 /50$$

RESULTS AND DISCUSSION

The contents of Pb, Cd, Co and Se in different common spices were presented in Table 2. The values of metal concentrations were compared with the maximum permissible concentration of 0.30, 0.2, and 3.50 mg kg⁻¹ for Pb, Cd, Co and Se respectively as recommended by Codex Alimentarius Commission (FAO/WHO 1984).

The lead contents of different samples are given in Table 2. As comparing with standard limit, the basibic sample has the highest content of lead (1.4 mg kg⁻¹) that far exceeds the standard level recommended by FAO/WHO (FAO/WHO 1984) (0.30 mg kg⁻¹). Samples of ginger, cardamon and thyme also contained higher concentrations of lead (0.4-0.9 mg kg⁻¹) than that recommended by FAO/WHO (FAO/WHO 1984). However, zero readings were obtained for turmeric, safflower, nutmeg, fenugreek, garden sage, mahaleb cherry, cinnamon, nasturinum, basibic, nigella, black pepper, cumin and coriander.

As shown in Table 2, the concentrations of cadmium of all the samples under investigation were under the maximum permissible concentration (0.20 mg kg⁻¹) of cadmium (FAO/WHO 1984). The amount of cadmium was in the range 0.04 mg kg⁻¹ in fenugreek and coriander to 0.14 mg kg⁻¹ for cardamon. This high level of cadmium might be due to the use of cadmium-containing phosphate fertilizers, or from the practice of growing these plants on soil amended with sewage sludge, or both. However, other samples like turmeric, garden sage, thyme, mahaleb cherry, cinnamon, and black pepper show no detectable amount of cadmium. These results may agree with what was

reported earlier (Waldraw and Stofen 1974) that lead concentration in food products ranged from undetectable levels to a few mg kg⁻¹ of wet weight.

Table 2 : Elements concentrations (mg kg⁻¹) on dry weight basis of studied commen spices

Spices name	Element (mg kg ⁻¹ on dry weight basis)			
	Lead	Cadmium	Cobalt	Selenium
Turmeric	Nil	0.10	0.32	4.40
Safflower	Nil	Nile	Nil	13.30
Nutmeg	Nil	0.03	Nil	Nil
Basisic	1.40	0.12	0.32	Nil
Nasturitum	0.20	0.12	0.32	Nil
Fenugreek	Nil	40	Nil	Nil
Garden sage	Nil	Nil	Nil	4.40
Thym	0.90	Nil	0.64	6.60
Mahlib	Nile	Nil	Nil	6.60
Cinnamon	Nil	Nil	Nil	2.20
Ginger	0.60	0.07	0.32	Nil
Cardamom	0.40	0.14	Nil	Nil
Nigella	Nil	Nil	Nil	4.40
Black Pipper	Nil	Nil	Nil	Nil
Cumin	Nil	0.08	Nil	4.40
Coriander	Nil	0.04	Nil	Nil
Standard limit	0.30	0.20	0.4	3.50

Varied levels of cobalt concentration were found as shown in Table 2. Samples of turmeric, megnut, fenugreek, garden sage, mahaleb cherry, cinnamon, cardamon, nigella, black pepper, cumin and coriander are almost free from cobalt. While the rest of samples contained variable amount of cobalt 0.32-0.64 mg kg⁻¹.

The levels of selenium are given in Table 2. The data shows variation in concentration of selenium for the investigated spices. Thus zero readings were obtained for nutmeg, basisic, nasturium, fenugreek, ginger, cardamon, black pepper, and coriander. The rest of samples contained amount in the range 2.2 mg kg⁻¹ in cinnamon to 13.3 mg kg⁻¹ in safflower. Other spices that exceed the recommended FAO/ WHO (1984) level included turmeric, safflower, garden sage, thyme, mahaleb cherry, nigella and cumin (3.50 mg kg⁻¹).

The results in Table 3 showed that no risk from daily intake of the most of spices under study for hazardous Pb, Cd, Co, and Se if the human intake is about 20 g of spices per day. But there are dangrous from basisic, thym and ginger for lead. Due to the high level of cadmium found in fenugreek therefor it could poisonous.

In conclusion, According to ATSDR (2001), the minimal risk levels for hazardous Pb, Cd, Co and Se through oral route and has acute effect are 0.0002, 0.0002, 0.01, 0.005 mg kg⁻¹day⁻¹ respectively. whereas the human needs from spices is very few grams per day there is no risk from used the species under study in the food. And also, there should be thorough control for imported food stuff at customs to meet FAO/ WHO recommendations and tolerable daily intake limits for heavy metals, and to avoid the passing for human consumption and prevent unknown disease.

Table 3 : Daily intake ($\text{mg kg}^{-1}\text{day}^{-1}$) more than 20 g of metals of studied common spices effect based on 50 g of human body.

Spices name	Risk duration for metal			
	Lead	Cadmium	Cobalt	Selenium
Turmeric	No effect	No effect	No effect	No effect
Safflower	No effect	No effect	No effect	No effect
Nutmeg	No effect	No effect	No effect	No effect
Basisic	Acute	No effect	No effect	No effect
Nasturitum	No effect	No effect	No effect	No effect
Fenugreek	No effect	acute	No effect	No effect
Garden sage	No effect	No effect	No effect	No effect
Thym	Acute	No effect	No effect	No effect
Mahlib	No effect	No effect	No effect	No effect
Cinnamon	No effect	No effect	No effect	No effect
Ginger	Acute	No effect	No effect	No effect
Cardamom	No effect	No effect	No effect	No effect
Nigella	No effect	No effect	No effect	No effect
Black Pepper	No effect	No effect	No effect	No effect
Cumin	No effect	No effect	No effect	acute
Coriander	No effect	No effect	No effect	No effect
Minimal risk levels	0.0002	0.0002	0.01	0.005

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REFERENCES

- Al-Eed, M. A., M.S. Al-Jasser and A. L. Selim. 1997. The chromatographic determination of fatty acids content and the chemical characteristics of some Saudi spices. *J. Agric. Sci. Mansoura. Univ.*, 22(5): 1685-1692.
- ATSDR .2001. Agency for toxic substances and disease registry. From web of <http://www.atsdr.cdc.gov/mrls.html>.
- FAO/WHO.1984. Joint FAO/WHO Food Standers Program, Codex Alimentarius Commission Contamination. CAC/Vol. XV11.FAO, Roma and WHO, Geneva.
- Geert, E., W. van Loon Johannes, and T. Kars. 1989. Heavy metals in vegetables grown in the Netherlands and in domestic and imported fruits. *Z Lebensm Unters Forsch* 190:34-39.
- Gilbert, J. 1984. Analysis of food contamination. Elsevier App. Sci. Pups., London 1.
- Husain, A., Z. Baroon, S. Al-Khalafawi, T. Al-Ati and W. Sawaya. 1995. Heavy metals in fruits and vegetables grown in Kuwait during the oil well fires. *Arab Gulf J. Sci. Research*. 13: (3)535-542.
- Kennish, M. J. 1992. Ecology of Estuaries. Anthropogenic effects. CRC. Press, Inc., Boca Raton, F1.
- Nitrates, Nitrites, and N-nitroso Compounds. Geeneva, World Health Organization, 1978 (Environmental Health Criteria 5).
- Oehme, F. W. 1989. Toxicity of heavy metals in the environment. Marcel Dekker, Inc., New York, Part 1, 1.
- Ozores-Hampton M., E. Hanlon, H. Bryan and B. Schaffer. 1997. Cadmium, copper, lead, nickel and zinc concentrations in tomato and squash grown in MSW compost-amended calcareous soil. *Compost Sci and Utilization*. 5: (4) 40-45.
- Sattar, A, M. Wahid and S.K Durrani. 1989. Concentration of selected heavy metals in spices, dry fruits and plant nuts. *Plant Foods For Human Nutrition*. 39(3) 279-286.
- Selim, A.I., M.S. Al-Jasser and M. A. Al-Eed. 1994. The fatty acids composition and the chemical characteristics of some umbelliferae spices. *Annals of Agric. Sc. Moshtohor*. 32(4): 1995-2004.

- Tsoumbaris, P. and H. Tsoukali-Papadopoulou. 1994. Heavy metals in common foodstuff: quantitative analysis. Bulletin of environmental contamination and toxicology. 53: part 1. pp. 61-66.
- Waldraw, H. A., Stofen, D. (1974) Sub-clinical lead poisoning. Academic Press, London
- Zakrzewski, S. F. 1991. Principle of environmental toxicology. ACS Professional reference book, Washington, DC, 1.

الملخص العربي

مستوي بعض من العناصر الثقيلة في البهارات الشائعة

محمد العيد، فهد السبيعي، محمد الجرواني ، هاني الهمشري ، الزين الطيب

قسم الكيمياء والنبات - كلية العلوم الزراعية والأغذية
جامعة الملك فيصل - ص ب ٤٢٠، الأحساء ٣١٩٨٢، المملكة العربية السعودية

قدرت كمية العناصر الثقيلة الرصاص والكاديوم والكوبالت و السلنيوم في بعض من البهارات الشائعة الأستعمال في الأسواق المحلية بالمملكة العربية السعودية [استخدام جهاز التقدير الذري. وقد وضحت الدراسة وجود فروق في تركيزات المعادن المختلفة طبقا للجزء المستعمل في الأكل (جذر، ساق، الأوراق، ثمار). تراوح تركيز الرصاص من كميات ضئيلة غير مقدره علي الجهاز الي ١٤,٣٠ مليجرام/كجم علي اساس الوزن الجاف بينما كان تركيز الكاديوم يختلف من ١,٢٥ مليجرام/كجم الي ٣,٠٥ مليجرام/كجم وكان تركيز الكوبالت من صفر الي ٠,٦٤ مليجرام/كجم بينما تراوح تركيز السلنيوم من صفر الي ١٣,٣ مليجرام/كجم. كانت بعض من هذه التركيزات فوق الحد المسموح به طبقا لمنظمتي الـ WHO و FAO . ليس هناك خطورة من تناول اليومي لجرعة ٢٠ جرام من لمعظم البهارات تحت الدراسة لشخص يزن أكثر من خمسين كيلوجرام بالنسبة لعناصر الرصاص ، الكاديوم، الكوبالت، السلنيوم ولكن خطورة حادة تظهر من تأثير الرصاص عند تناول ٢٠ جرام من أنواع الحبك أو الزعتر و الزنجبيل بينما خطورة الكاديوم تظهر عند تناول نفس الجرعة في اليوم من الحلبة.

