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Full Length Research Paper

Lead and cadmium residue determination in spices available in Tripoli City markets (Libya)

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In recent years, there has been a growing interest in monitoring heavy metal contamination in food products. Spices can improve the taste of food and can also be a source of many bioactive compounds but can unfortunately, also be contaminated with dangerous materials, potentially heavy metals. This study was conducted to investigate lead (Pb) and cadmium (Cd) contamination in selected spices commonly consumed in Libya including *Capsicum frutescens* (chili pepper), *Piper nigrum* (black pepper), *Curcuma longa* (turmeric) and mixed spices (HRARAT) which consist of a combination of: *Alpinia officinarum, Zingiber officinale* and *Cinnamomum zeylanicum*. Spices were analyzed by atomic absorption spectroscopy after digestion with nitric acid/hydrogen peroxide. The highest levels of lead (Pb) was found in *Curcuma longa* and *Capsicum frutescens* in wholesale markets (1.05 ± 0.01 mg/kg, 0.96 ± 0.06 mg/kg). Cadmium (Cd) levels exceeded FAO/WHO permissible limit. *C. longa* and *P. nigrum* sold in retail markets had a high concentration of Cd (0.36 ± 0.09, 0.35 ± 0.07 mg/kg, respectively) followed by 0.32 ± 0.04 mg/kg for *C. frutescens*. Mixed spices purchased from wholesale markets also had high levels of Cd (0.31 ± 0.08 mg/kg). *C. longa* and *C. frutescens* may pose a food safety risk due to high levels of lead and cadmium. Cadmium levels exceeded FAO/WHO recommendations (0.2 ppm) for *P. nigrum, C. alonga* and HRARAT.

Key words: Heavy metals, lead, cadmium determination, spice, Libya.

INTRODUCTION

In the last decade, interest has grown concerning the dangerous effects of heavy metals on human health resulting from environmental pollution and the prevalence of heavy metals in trace food components such as spices that could result from environmental exposure from the atmosphere, soil and water that eventually find its way into food creating a health risk for humans and animals (Kabata-Pendias, 2011). These toxic metals reach agricultural crops during cultivation, or through industrial activities such as mining, from industrial waste, waste water, pesticides and packaging material (Bradl, 2005; Sarpong et al., 2012; Siegel, 2002). There are several metals of particular concern: lead (Pb), cadmium (Cd), tin (Sn), arsenic (As) and mercury (Hg). Cadmium and lead are

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among the most toxic (Siegel, 2002; Sullivan et al., 2007) due in part to the fact that they accumulate in biological tissues and increase from lower to higher trophic levels, a phenomenon known as biomagnification (Sullivan et al., 2007). Heavy metal damages human health in two ways. The first is disruption of normal cellular processes leading to toxicity. The second, particularly for cadmium and lead is bioaccumulation, particularly in the liver or kidney, where these metals are excreted at a slower rate as compared to uptake (Apostoli, 2006; Zelikoff and Thomas, 1998).

Important food sources of toxic metals are plant foods including spices. Spices are derived from buds, barks, rhizomes, fruits, seeds and other parts of the plant (Peter, 2001). Spices are responsible for making food dishes more distinctive, palatable and aromatic and may contain toxic metals derived from the surroundings through production, processing and marketing (Inam et al., 2013; Ziyaina, 2007). The use of spices and herbs has increased markedly in most regions of the world, including Europe and North America (Nkansah, 2010) with importation from South Asia and developing countries increasing. Several recent food borne incidents have involved both intentionally and unintentionally adulterated spices and herbs with heavy metals added to improve color. Contaminated spices are dumped onto markets in developing countries that have limited ability to test for adulterants. Due to the risk, it is important to evaluate the levels of lead and cadmium in milled spices (red pepper, black pepper, turmeric and mixed spices (HARART) that are commonly consumed in Libya and have a history of adulteration and also to determine the sources and distribution of these metal contaminants in milled species.

Libya, outside of Egypt, is the largest market for spices in North Africa. Central markets in Tripoli serve the entire region from Algeria to Tunisia and into Chad and countries further south and west. India commands 40% of the world chilli market, 11% of the turmeric market and 5% of the black pepper market (www.marketsandmarkets.com). Much of the *Capsicum* spp. and *Piper* spp. sold in Libya is sourced from India, Pakistan and Turkey. High levels of contamination with toxins and filth have been previously reported (fda.gov) possibly due from contaminated irrigation water, and as a result of these perceived risks, a survey was conducted in the Libyan market to assess potential food safety risk.

The objective of this study was to determine the prevalence of lead and cadmium in selected ground spices available in Libyan retail markets.

MATERIAL AND METHODS

Sample collection

Table 1 lists the spices and the part of the plant used. Some of the most consumed spices in Libya include *Capsicum frutescens*, *Piper nigrum, Curcum alonga* and mixed spices (Ziyaina, 2007). The study focused on the contamination assessment of spices that are imported and traded within Libyan markets in 2011. Twenty four samples of each type of spice were collected upon the arrival of these spices to Libya from seven wholesale markets, which are the main sources of spices entering into the country. An additional 36 samples for each type of spice were collected from several retailers in metropolitan Tripoli, Libya.

Sample preparation

Homogenized spice samples were dried in an oven at 100°C for 24 h and then 5 g was accurately weighed into a beaker. Concentrated nitric acid (HNO_3) (65%) was added (5.0 ml) followed by 2.5 ml of 30% hydrogen peroxide (H_2O_2). Samples were left at room temperature for a few minutes and then heated on an electric heater (120°C) and mixed. Five (5) ml of 65% HNO_3 was added and the digests reheated (120°C, few minutes), followed by the addition 10 ml of distilled water. Sample digests were then filtered through Whatman No. 42 filter paper and <0.45 ml and diluted to volume (Yash, 1998). Pb (217 nm) and Cd (228.8 nm) were determined by atomic absorption spectrometry (Varian AAS240, USA). The standard solution for analyses and development of a calibration curve was prepared by diluting a stock solution of 1000 mg L^{-1} of the examined heavy metals. All chemicals used in this experiment were from Sigma Aldrich, St. Louis MO, USA).

Statistical analysis

Means and standard deviations were computed using SAS.LMC. statistical software (SAS Institute Inc, Cary, NC USA) and the Duncan's multiple range test (MRT).

RESULTS AND DISCUSSION

The range and average Pb and Cd in spices from two sources (retailer and wholesale markets) are presented in Tables 2 and 3.

In retail markets, a significant ($P<0.01$) number of spices contained lead. *C. longa* and *C. frutescens* from wholesale markets had the highest levels of contamination. Mixed spices had the lowest concentration of lead, but the levels were still relatively high (0.65 ± 0.09 mg/kg). These values were below the maximum permissible level (10 ppm) recommended by FAO/WHO (2006). Nevertheless, it is important to take the necessary steps to perform routine monitoring of the levels of lead in these spices in order to avert a public health risk since high levels of Pb have been found in other studies on herbs and spice plants from different parts of the world (Seddigi et al., 2013). Chizzola and others (2003) found that heavy metals including Pb were generally within an acceptable range in herbs, spices and medicinal plants on Austrian markets (Chizzola et al., 2003). On the other hand, studies conducted in Poland found the average lead content to be about 1.49 mg/kg in cinnamon, which exceeds the maximum permissible level (Krejpcio et al., 2007).

As shown in Table 2, mixed spices sold in retail markets have a high concentration of Cd (0.36 ± 0.09 mg/kg). The Cd content in black pepper, turmeric and mixed spices were over the maximum permissible limit

Table 1. The names of spices surveyed.

Scientific name	Commercial name	Part used
Capsicum frutescens	Red Pepper	Fruits
Piper nigrum	Black pepper	Seeds
Curcuma longa	Turmeric	Rhizome
Mixed spices		
Alpiniaofficinarum	Galangal	Rhizome
Zingiberofficinale	Ginger	Rhizome
Cinnamomumzeylanicum	Cinnamon	Bark

Table 2. Concentration of lead (Pb) in spices from Libyan markets.

Spice	Element (mg/kg)	Source	Max.	Min.	Average ± S.D
Capsicum frutescens	Pb	Wholesale	0.95	0.81	0.88 ± 0.07
		Retailer	1.02	0.90	0.96 ± 0.06
Piper nigrum	Pb	Wholesale	0.80	0.66	0.73 ± 0.07
		Retailer	0.90	0.69	0.82± 0.13
Curcuma longa	Pb	Wholesale	0.70	0.56	0.63 ± 0.01
		Retailer	1.06	0.96	1.005 ± 0.01
*Mixed spices	Pb	Wholesale	1.00	0.84	0.92 ± 0.08
		Retailer	0.74	1.04	0.89 ± 0.09

*Mixed spices: *Alpinia officinarum, Zingiber officinale* and *Cinnamomum zeylanicum.*

Table 3. Concentration of cadmium (Cd) in spices from Libyan markets.

Spice name	Element (mg/kg)	Source	Maximum limit	Minimum limit	Average ± S.D
Capsicum frutescens	Cd	Wholesale	0.17	0.06	0.14 ± 0.01
		Retailer	0.22	0.16	0.19 ± 0.08
Piper nigrum	Cd	Wholesale	0.19	0.11	0.15 ± 0.07
		Retailer	0.39	0.25	0.32 ± 0.04
Curcuma longa	Cd	Wholesale	0.24	0.14	0.19 ± 0.08
		Retailer	0.39	0.32	0.35 ± 0.07
Mixed spices	Cd	Wholesale	0.40	0.22	0.31 ± 0.08
		Retailer	0.51	0.42	0.36 ± 0.09

(0.2 ppm) recommended by FAO/WHO 2006 (Table 2). The average values of Cd were close to those found in a study by Bempah (2012) that reported the highest concentration of Cd to be 0.44 mg/kg in *Ocimum viride*. The values found here for Libyan markets are high, but lower than what has been found in other markets. In one study in India, the average concentrations of Cd in medicinal plants and spices ranged from 0.684 to 2.751 mg/kg (Subramanian et al., 2012). Other studies have found high Cd concentrations in *Piper nigrum* (206 mg kg^{-1}) and cinnamon *(*124 mg kg^{-1}). Cadmium concentrations in medicinal plants were variable, but often high: in Italy (10-750 mg kg^{-1}), Egypt (50-300 mg kg^{-1}) and Brazil (<0.2-0.74 mg kg^{-}) (Abou-Arab et al., 2000; Caldas et al., 2004).

In general, heavy metal content in spices reflects

environmental pollution levels, bioaccumulation in plant tissue, application of lead or heavy metal containing materials such as arsenate based pesticides (Krejpcio et al., 2007; Nkansah, 2010). High levels of heavy metals could be due to the use of heavy metal-containing fertilezers or from a practice of growing plants with sewage sludge (Ibrahim et al., 2012; Inam et al., 2013).

Conclusion

The levels found in this study for lead in *C. longa, C. frutescens*, and mixed spices (*Alpinia officinarum, Zingiber officinale* and *Cinnamomum zeylanicum*) were below those recommended by FAO. However, levels of cadmium exceeded FAO recommendations for *P. nigrum*, *C. longa*, and mixed spices. Differences observed between Cd and Pb levels for spices sold in retail and wholesale markets indicate that the quality of spices across the value chain in Libya is highly variable and that a number of sources supply the market, some of which are contaminated and some of which are not.

Further studies should be conducted to estimate intake of these and other spices by consumers in the Libya and regionally where a similar cuisine is eaten and where there is a similar lack of control on imported ingredients to ascertain whether there is a health risk.

Conflict of Interests

The author(s) have not declared any conflict of interests.

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